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(71)Applicant : TOKUYAMA CORP

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(72)Inventor : WATANABE SHIN
SAKAKI TORU

(54) SUBSTANCE MEASURING UTENSIL, ITS MANUFACTURE AND ION MEASURING UTENSIL

(57)Abstract:

PURPOSE: To analyze and extract a substance in a solution simply and in a short time by a method wherein a water-soluble polymer phase is filled into a part between organic phases having a three-dimensional network structure which is formed by dissolving a non-water-soluble polymer in a hardly-water-soluble volatile organic solvent.

CONSTITUTION: A non-water-soluble polymer such as a polyvinyl chloride-based polymer, a styrene-based polymer or the like is dissolved in a hardly-water-soluble volatile organic solvent (an organic solvent) such as dioctyl phthalate, o-nitrophenyloctyl-ether or the like, and organic phases having a three-dimensional network structure whose viscosity and physical strength are suitable for use as a substance-measuring utensil are generated. A water-soluble polymer such as polyethylene glycol or the like is filled into a part between the organic phases so that a sample to be measured is permeated quickly into the part between the organic phases. Although various filling methods are available, a method in which, after the organic solvent, the non-water-soluble polymer and the water-soluble polymer have been dissolved in the organic solving medium, they are coated on a support so as to be dried is most suitable.

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CLAIMS DETAILED DESCRIPTION TECHNICAL FIELD PRIOR ART EFFECT OF THE
INVENTION TECHNICAL PROBLEM MEANS EXAMPLE DESCRIPTION OF DRAWINGS
DRAWINGS

[Translation done.]

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CLAIMS

[Claim(s)]

[Claim 1] (A) The matter measurement implement with which it consists of the organic phase and (B) water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule in a difficulty water solubility difficulty volatility organic solvent, and (A) organic phase is characterized by (B) water soluble polymer phase being filled up with three-dimension-network structure by the nothing (** A) organic interphase.

[Claim 2] The matter measurement implement with which it comes to carry out the laminating of the matter measurement implement according to claim 1 on a base material.

[Claim 3] The manufacture approach of the matter measurement implement according to claim 2 characterized by evaporating an organic solvent under the condition which the solution which comes to dissolve a difficulty water solubility difficulty volatility organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer in an organic solvent is applied on a base material, and the deposit of the organic phase which subsequently consists of a difficulty water solubility difficulty volatility organic solvent and a nonaqueous solubility macromolecule starts before a deposit of a water soluble polymer, or deposits in coincidence.

[Claim 4] (A) The coloring matter which carries out a mutual reaction and shows a detectable response to the ionophore and this complexing which carry out complexing to the ionophore or ion which carries out complexing to ion and shows a detectable response to a difficulty water solubility difficulty volatility organic solvent, The ion measurement implement with which it consists of the organic phase and (B) water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule in a list, and (A) organic phase is characterized by (B) water soluble polymer phase being filled up with three-dimension-network structure by the nothing (** A) organic interphase.

[Claim 5] The ion measurement implement with which it comes to carry out the laminating of the ion measurement implement according to claim 4 on a base material.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the ion measurement implement which used this for the matter measurement implement list which can be used for detection of various matter in a solution, and a quantum.

[0002]

[Description of the Prior Art] In the field of analytical chemistry, measuring various matter especially in a water solution, and using for medical care, a production activity, environmental protection, etc. is performed among the solution. For example, it is known by measuring the ion concentration in the blood serum of the body, and urine that the useful information about a patient's symptoms will be obtained. Moreover, measurement of the concentration of the surfactant contained in the monitor of the harmful heavy metal in the waste fluid discharged as a result of the production activity of an enterprise, domestic wasted water, etc. is needed for environmental protection. In addition, although there will be no enumeration if an example is given, measurement of the matter in a solution is not dispensable to our life.

[0003] The measuring method of the matter in a solution changes with the measuring object matter, and many approaches exist. There is the approach of calling a colorimetric method which measures the absorption by which induction is carried out with the measuring object matter into it, or change of a fluorescence spectrum. In a colorimetric method, extraction separation of the measuring object matter in a water solution is carried out into an organic solvent, and a quantum is performed in many cases from the variation of the absorption produced by the interaction of the measuring object matter which moved to the organic solvent, and the color reagent beforehand added underwater or in the organic solvent, or a fluorescence spectrum.

[0004] Extraction separation is actuation to which a certain matter is made to separate into another side from one side of two sorts of liquids and which is performed for accumulating. Actuation of taking out only the near solution which two sorts of solutions which generally are not mixed mutually were put into the separating funnel, this was shaken, and the measuring object matter moved after that is performed. Therefore, in measurement of the colorimetric method for which extraction separation actuation is needed, the tool of dedication for extraction separation is needed. In order to compensate this fault, using the film which consists of a difficulty water solubility difficulty volatility organic solvent which added the nonaqueous solubility macromolecule instead of the organic solvent is known. In volume [40th] (1991) 227-231 pages for example, "analytical chemistry" — The film made to dissolve the bathocuproine which forms a complex specifically with the copper ion of monovalence, and is colored in a nonaqueous solubility macromolecule and a difficulty water solubility difficulty volatility organic solvent is used. Carry out extraction separation of the copper ion into the film, the bathocuproine in the film is made to color, and the approach of measuring copper ion concentration from a membranous absorbance is described by by contacting this film in the water solution containing the copper ion of the monovalence of strange concentration. However, since the mass transfer that the measuring object matter in a water solution moves into a difficulty water solubility difficulty volatility organic solvent in extraction separation actuation

was indispensable, diffusion in the difficulty water solubility difficulty volatility organic solvent of the measuring object matter became rate-limiting, and there was demerit in which the time amount concerning measurement was very long.

[0005] The analysis method of the ion by the colorimetric method of the principle which produces absorption or change of a fluorescence spectrum by migration of the measuring object matter is stated to JP,59-211864,A, JP,59-231452,A, and JP,60-194360,A. These are ion concentration measurement implements which consist of a difficulty water solubility difficulty volatility organic solvent containing various ionophores and the coloring matter called the reporter matter. In these measurement implements, by making the test portion water solution containing the ion which is the measuring object matter contact, migration into the film of ion breaks out and change is produced in the extinction spectrum or fluorescence spectrum of coloring matter. It is known that the rate which ion diffuses within the film also in this approach will become rate-limiting. It is known that it is effective technique to make thickness thin to about several micrometers. However, while it becomes large, since the rate of coloring change has the fault that the coloring variation obtained becomes small, it is not practical.

[0006] Various technique has been developed in order to conquer the above-mentioned fault. For example, the ion concentration measurement implement which made porosity support support the difficulty water solubility difficulty volatility organic solvent containing pH susceptibility coloring matter called various ionophores and the reporter matter to JP,60-194360,A is indicated. It was the attempt in which both the rate of coloring change and the problem of coloring variation would be solved, by making the front face of porosity support like a filter paper cover thinly an ionophore and the difficulty water solubility difficulty volatility organic solvent containing the reporter matter. A certain amount of solution was able to be carried out by this technique. However, when the measurement implement using these porosity support manufactured, it needed the process of fixing porosity support on a base material in an exact location, and in addition, manufacturing to a large quantity and homogeneity had difficulty or the fault that cost started dramatically. Therefore, development of technique with the effectiveness same [without using porosity support] was desired.

[0007]

[Problem(s) to be Solved by the Invention] When it is used for the matter measurement implement which can perform measurement which combines practically sufficient speed of response and the amount of responses, for example, a colorimetric method, it aims at development of the matter measurement implement which can perform measurement which combines practically sufficient coloring rate and coloring variation, and the ion measurement implement which used this for the list.

[0008]

[A means to solve a technical problem] this invention persons repeated research wholeheartedly that the matter measurement implement which can solve this technical problem should be developed. Consequently, when the organic phase and water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule in a difficulty water solubility difficulty volatility organic solvent took specific structure, it found out demonstrating sufficient engine performance solving the above-mentioned technical problem.

[0009] That is, this invention is (A). It consists of the organic phase and (B) water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule in a difficulty water solubility difficulty volatility organic solvent, (A) organic phase is the matter measurement implement characterized by (B) water soluble polymer phase being filled up with three-dimension-network structure by the nothing (** A) organic interphase, and it is the matter measurement implement with which it comes to carry out the laminating of this matter measurement implement on a base material further.

[0010] Other invention is (A). Coloring matter which carries out a mutual reaction and shows a detectable response to the ionophore and this complexing which carry out complexing to the ionophore or ion which carries out complexing to ion and shows a detectable response to a difficulty water solubility difficulty volatility organic solvent, It consists of the organic phase and (B) water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule

in a list. (A) An organic phase is the ion measurement implement characterized by (B) water soluble polymer phase being filled up with three-dimension-network structure by the nothing (** A) organic interphase, and it is the ion measurement implement with which it comes to carry out the laminating of this ion measurement implement on a base material further.

[0011] Furthermore, other invention, It is the manufacture approach of the above-mentioned matter measurement implement characterized by evaporating an organic solvent under the condition which the solution which comes to dissolve a difficulty water solubility difficulty volatility organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer in an organic solvent is applied on a base material, and the deposit of the organic phase which subsequently consists of a difficulty water solubility difficulty volatility organic solvent and a nonaqueous solubility macromolecule starts before a deposit of a water soluble polymer, or deposits in coincidence.

[0012] The organic phase which constitutes the matter measurement implement of this invention consists of a difficulty water solubility difficulty volatility organic solvent and a nonaqueous solubility macromolecule fundamentally.

[0013] The above-mentioned difficulty water solubility difficulty volatility organic solvent (only henceforth an organic solvent) points out the thing of an organic compound liquefied in the ordinary temperature which does not have the functional group which is a high-boiling point and can be ionized refractory to water. This organic solvent is used in order to extract the measuring object matter in a test portion and to dissolve coloring matter required for measurement etc. The boiling point is specifically 100-degree more than Centigrade, and it is desirable in order to carry out measurement by which it was stabilized that the organic solvent which can dissolve to 1l. of water is 1g or less extent.

[0014] If the organic solvent used suitably is mentioned concretely, there is high-boiling point ether, such as high-boiling point ester, such as adipic-acid dibutyl and a dioctyl phthalate, o-nitrophenyl octyl ether, and diphenyl ether.

[0015] As a nonaqueous solubility macromolecule, it can dissolve in the above-mentioned organic solvent, and an insoluble macromolecule is used for water. This nonaqueous solubility macromolecule dissolves in the above-mentioned organic solvent, and gives the viscosity and physical reinforcement suitable for the activity as a matter measurement implement.

[0016] If the example of the nonaqueous solubility giant molecule used suitably is given, there are styrene system polymers, such as methacrylic ester system polymers, such as vinylidene-chloride system polymers, such as polyvinyl chloride system polymers, such as a polyvinyl chloride and an ethylene-vinyl chloride copolymer, a vinyl chloride-vinylidene-chloride copolymer, and a vinylidene chloride, and a polymethyl methacrylate, and polystyrene, etc. Although it can be used without being limited especially about the polymerization degree of a nonaqueous solubility macromolecule, if a with an average degree of polymerization of 500 or more thing is generally used, an organic solvent comes to show free-standing, and the handling when considering as a matter measurement implement it being the range where it is below the upper limit of the solubility, and an organic solvent shows free-standing becomes easy and is suitable for the concentration in the organic solvent of a desirable nonaqueous solubility macromolecule. Although the density range changes with classes of the nonaqueous solubility macromolecule to be used and organic solvent, it is suitable to make it dissolve five to 100% of the weight, and to use a nonaqueous solubility macromolecule to an organic solvent, generally. Especially the dissolution approach to the organic solvent of a nonaqueous solubility macromolecule is not limited, but it is dissolved, usually adding a nonaqueous solubility macromolecule gradually into an organic solvent.

[0017] The water soluble polymer phase which constitutes this invention consists of a water soluble polymer fundamentally. This water soluble polymer is used in order to promote that a test portion permeates between the organic phases of three-dimension-network structure promptly.

[0018] A water soluble polymer is a macromolecule which can dissolve in water, and the solubility is [a water soluble polymer] 50 wt%. It is desirable to use the above thing. If a water soluble polymer does not exist, since can go a test portion and it cannot be crossed even to all

the corners of an organic solvent, the measuring time required when a matter measurement implement is used for measurement will become long. It is desirable to use a thing refractory to an organic solvent for a water soluble polymer. Even if it uses what is dissolved in an organic solvent, the effectiveness of this invention can be acquired, but when contacting the matter measurement implement of a test portion and this invention in that case, stable measurement may not be able to be performed with remarkable deformation. Moreover, when the ease of the handling at the time of using it as a matter measurement implement is taken into consideration, it is desirable to use the thing of the polymerization degree which will be in a solid state in a room temperature. Maintaining suitable physical reinforcement to the amount of whole including an organic solvent and a nonaqueous solubility macromolecule, if the amount of a water soluble polymer is 50 – 100% of the weight of the range, although osmosis of a test portion is promoted, it is desirable.

[0019] If the example of the water soluble polymer which can be used suitably is given, there are polyether system high molecular compounds, such as a polyethylene glycol and a polypropylene glycol, a polyvinyl pyrrolidone, polyvinyl alcohol, polyacrylamide, etc.

[0020] It can check that the organic phase of this invention is three-dimension-network structure by observing the part which remains after flushing a water soluble polymer with water with an electron microscope etc.

[0021] Although various approaches can be used for the approach of making a water soluble polymer phase existing in the organic interphase of three-dimension-network structure in the state of restoration, the approach used most suitably is as follows.

[0022] That is, it is made to dissolve in an organic solvent, and all of an organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer are applied on a base material after that, and they are dried. The micro phase separation of an organic phase and a water soluble polymer phase which comes to dissolve a nonaqueous solubility macromolecule in an organic solvent occurs with evaporation of an organic solvent, and where the organic phase of three-dimension-network structure is filled up with a water soluble polymer phase, it deposits.

[0023] If all of an organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer are dissolved as an organic solvent used for the above-mentioned approach, it can use that there is no limit in any way. Although the organic solvents which can be used with the combination of the selected organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer differ, if the example is given, there are alkyl halides, such as ether, such as a tetrahydrofuran, dichloromethane, chloroform, and a dichloroethane. Since the amount of the organic solvent used changes with solubility of the selected organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer, it cannot generally specify, but in order to urge a deposit, it is common to carry out an organic solvent, a nonaqueous solubility macromolecule, or a water soluble polymer to an organic solvent and more than tales doses.

[0024] By the above-mentioned depositing method, when the deposit of the organic phase which consists of an organic solvent and a nonaqueous solubility macromolecule starts before the deposit of a water soluble polymer phase or it deposits in coincidence, uniform and good structure can be acquired. The case where it manufactures on the conditions from which the deposit of an organic phase begins previously is more suitable. On the contrary, when a deposit of a water soluble polymer starts before the deposit of an organic phase, the matter measurement implement obtained becomes an ununiformity and what has many openings, and it is not suitable for measurement by the colorimetric method.

[0025] Control of the deposit sequence of an organic phase and a water soluble polymer phase can be carried out by controlling the solubility over the organic phase and water soluble polymer phase of the organic solvent in which a bi-phase is dissolved. Generally this soluble control is controlled by the polarity of an organic solvent. Being easy to dissolve the organic solvent and nonaqueous solubility macromolecule which generally constitute an organic phase in a polar low organic solvent, a water soluble polymer has conversely the property to be easy to dissolve in a polar high organic solvent. Therefore, if a polar high organic solvent is used, the deposit of an organic phase with low solubility will start previously. Therefore, the deposit of the organic phase which consists of an organic solvent and a nonaqueous solubility macromolecule can be started

before the deposit of a water soluble polymer phase.

[0026] The polarity of an organic solvent is controllable by mixing two or more sorts of organic solvents or water. If the example used suitably is shown, there are a tetrahydrofuran which added about 0.1% of water, chloroform which added the methanol 1%. Moreover, also when an ionophore and coloring matter are dissolved in an organic phase, the sequence of a deposit can be controlled by the same approach of being satisfactory at all.

[0027] If the concrete example which obtains the matter measurement implement of good structure is given, as an organic solvent, a polymethyl methacrylate can be used as o-nitrophenyl octyl ether and a nonaqueous solubility giant molecule, a polyethylene glycol (average molecular weight 6000 [about]) can be used as a water soluble polymer, these can be dissolved in a tetrahydrofuran, and it can apply on a glass plate, and can produce by making it leave and dry in a room temperature. If the moisture of a minute amount exists in a tetrahydrofuran at this time, a deposit of a polyethylene glycol becomes slow and can acquire the above-mentioned structure with sufficient repeatability. By using the organic phase in which the organic colored phase or coloring matter was dissolved, when the colored sludge deposits, it can know that the deposit of the organic phase which consists of an organic solvent and a nonaqueous solubility macromolecule will start before the deposit of a water soluble polymer phase.

[0028] the principle and approach of using for measurement of ion concentration the organic phase which comes to dissolve the ionophore which carries out complexing to ion and shows a detectable response to an organic solvent — well-known — “analytical chemistry” — volume [40th] (1991) 227–231 pages have the description. Moreover, the principle and approach of using for measurement of ion concentration the coloring matter which carries out a mutual reaction and shows a detectable response to the ionophore and this complexing which carry out complexing to ion at an organic solvent, and the organic phase which comes to dissolve a nonaqueous solubility macromolecule in a list are also well-known, and JP,59-211864,A, “analytical Science” (ANALYTICAL SCIENCE) 1990 year 715–720 page, etc. have the description. The ionophore and coloring matter which are used in this invention can use satisfactory at all what is stated by these reference.

[0029] In this invention, an ionophore is a compound with the property which carries out a specific cation and specific complexing. The coloring matter which has the capacity which forms a specific metal ion and a specific complex, produces change in the electron orbit condition by forming this metal ion and a complex, and produces change in the absorption spectrum or fluorescence spectrum of a complex can be used as an ionophore which carries out complexing to the ion of this invention, and shows a detectable response. As such an ionophore, there are nitroso aminophenols, diethylamino phenols, porphyrins, and phenoxazine.

[0030] Moreover, as an ionophore which can be used in the combination of the coloring matter which carries out a mutual reaction and shows a detectable response to the ionophore which carries out complexing to the ion of this invention, and the complexing concerned, there are several sorts of well-known cyclic peptide compounds, annular or an un-annular polyether derivative, a POTANDO derivative, a calyx allene derivative, etc.

[0031] If the example which can be used suitably [the ionophore which forms ion and a complex and shows a detectable response] is given potassium ion — receiving — 3 — ‘- nitro-4’ - (2, 4, 6-trinitro phenylamino) benzo-18-crown -6 and 4’ - (2, 6-dinitro-4-trifluoro methylphenyl) amino benzo-15-crown-5 grade To a copper ion, bathocuproine etc. receives iron ion. Nitroso aminophenols, such as 2-nitroso-5-N and N-dipropylamino phenol As opposed to nickel and zinc ion diethylamino phenols, such as a 2-(5-BUROMO-2-pyridyl azo)-5-diethylamino phenol To a hydrogen ion, there are azo phenols, such as phenoxazine, such as [9-(diethylamino)-5-octadecanoyl imino-5H-[benzoa] phenoxazine], and a [5-OKUTADEKANO yloxy-2-(4-nitro phenylazo) phenol].

[0032] If the example which can be used suitably [the ionophore used for the ionophore which carries out complexing to ion and the complexing concerned on the other hand in the combination of the coloring matter in which a mutual reaction is carried out and a detectable response is shown] is given As a lithium ionophore, it is 6 and 6-dibenzyl. - 1, 4, 8, 11-tetra-OKISA cyclo tetradecane, 2, the 9-dibutyl -1, 10-phenanthroline, etc. are 5, 11, 17, and 23-

tetra-*t*-butyl as a; sodium ionophore. — It is 25, 26, 27, and 28. — Tetrakis (ethoxycarbonyl)-methoxy-calyx [4] allene, Screw [(12-crown -4) methyl] methyl dodecyl malonate etc. as a; potassium ionophore Valinomycin, 2, 3-naphth - 1, 4, 7, 10, 13-PENTAOKISA cyclo PENTADEKA-2-en, Screw [(benzo-15-crown -5) -4'-methyl] PIMERETO, Valinomycin etc. as a; calcium ionophore ** — diethyl - N — N — ' — [(4R, 5R) - four — five - dimethyl - one — eight - dioxo - three — six - dioxa — octamethylene —] — a screw (12-methyl friend throat decanoate) — etc. — as; magnesium ionophore — The benzo-15-thia crown-4-ether etc. has ** N, N'-diheptyl-N, an N'-dimethyl ASUPARUTO amide, etc. to a copper ion.

[0033] Of course, in addition to this, the compound with which forming a cation and a complex is known exists, and can also use them. As reference about an ionophore, the volume Yoshihisa Inoue and on George W.Gokel "Cation Binding by Macrocycles" (MARCEL DEKKER INCORPORATION 1990), 2013 — 2016 pages (ANALYTICAL CHEMISTRY) (1988) of work "application to analytical chemistry of functional macrocyclic compound" (IPC, Inc. 1990 issuance) 60-volume Analytical Chemistry edited by Hiroyuki Takeda, "Analytical chemistry" 23 pages [1412 — 1430 etc.] (1974) etc. are mentioned.

[0034] There is a series of coloring matter called a pH indicator or pH susceptibility coloring matter as coloring matter (it is henceforth described as reporter coloring matter) which carries out a mutual reaction and shows a detectable response to the ionophore and this complexing which carry out complexing to above-mentioned ion.

[0035] Since the reporter coloring matter used in this invention is solubilized by the organic solvent and contained, what has it is used. [remarkable water solubility and small] The reporter coloring matter of 5-10 is suitably used for electric dissociation exponent from extent of the coloring response when using it as the ease of the acquisition, and an ion measurement implement.

[0036] If reporter coloring matter is shown concretely, there are triphenylmethane dye derivatives, such as indophenol derivatives, such as a phenolphthalein derivative [, such as a phenolphthalein and a tetrabromo phenolphthalein,], 2, 6-dichloroindophenol, 2, and 6-dibromo-3'-methyl indophenol, and Victoria pure blue, etc.

[0037] An ionophore and reporter coloring matter can be dissolved in an organic solvent to the upper limit of the solubility. At this time, it is known that it is more suitable for the mole ratio of an ionophore and reporter coloring matter than that principle that it is 1:1. Generally, the concentration in the inside of the organic solvent of an ionophore and reporter coloring matter takes the sensibility when measuring into consideration, and it is desirable that it is the range of 0.1-200mM, respectively.

[0038] In this invention, it points out that the organic phase has network structure in three dimension like sponge and sponge as the organic phase which comes to dissolve a nonaqueous solubility giant molecule in an organic solvent is three-dimension-network structure. As for the matter with the same structure, various things are used with different construction material as the absorptivity matter or an object for filtration separation. The process of the matter which consists of a nonaqueous solubility macromolecule with three-dimension-mesh-like structure is indicated by JP,2-302449,A, JP,58-104940,A, JP,47-4593,A, etc. The matter of such conventional three-dimension-network structures makes a nonaqueous solubility macromolecule a subject, in order to realize high reinforcement, and generally it does not add an organic solvent. Moreover, even if the organic solvent is added, what was filled up with the water soluble polymer is not known. The organic phase which comes to dissolve a nonaqueous solubility macromolecule in the organic solvent of the three-dimension-network structure of this invention makes a subject the organic solvent for performing extract operation, and has a fundamental difference that a nonaqueous solubility macromolecule is added in order to give moderate viscosity and physical reinforcement.

[0039] In this invention, a test portion permeates in the micropore of the organic phase of three-dimension-network structure promptly by filling up the organic interphase of three-dimension-network structure with the water soluble polymer phase. since [moreover,] an organic phase is three-dimension-network structure — several micrometers or less — thickness can be carried out, consequently migration into the organic phase of the measuring object matter can be

completed promptly. Moreover, the aperture of the micropore made when the water soluble polymer of the organic phase of three-dimension-network structure is removed can take uniform structure, when it is 500 micrometers or less. By taking uniform structure, more exact measurement is enabled and it is desirable.

[0040] Although the matter measurement implement of this invention can be used for measurement even if it remains as it is, it can take into consideration an activity in combination with reagents, such as operability in the case of measurement, and a buffer for pH, etc., and it can carry out a laminating on a base material. A well-known approach can be used without limiting especially as the approach of a laminating. How to produce the matter measurement implement of this invention beforehand, and paste up with suitable adhesives, a double-sided tape, etc., if the example used suitably is given. The solution which comes to dissolve an organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer in an organic solvent can be applied on a base material, and the organic phase which comes to dissolve a nonaqueous solubility macromolecule in an organic solvent as above-mentioned can measure concentration of various matter using the matter measurement implement with the approach of drying under the conditions which deposit in three-dimension-network structure etc. which carried out in this way and was produced. It explains to below with an example.

[0041] Measurement of the concentration of the surfactant of the surfactant in current and a water solution adds specifically association or the coloring matter which carries out complexing in a water solution with a surfactant, extracts both a surfactant and the coloring matter to the extract of nonaqueous solubility out of a water solution, and is performed by measuring whenever [coloring / of an extract] after that. Such an approach is shown in Japanese Industrial Standards (JIS K 0102-1986 testing method for industrial waste water), and the methylene-blue absorptiometry, the tetra-thio cyano cobalt (II) acid absorptiometry, etc. are indicated. After each of these approaches puts two kinds of liquid into a container, in order to accompany them by actuation of shaking and extracting, they has the fault of requiring serious time amount and a serious effort.

[0042] The above-mentioned measurement can be performed by the same principle using the matter measurement implement of this invention. It will be as follows if a methylene-blue absorptiometry is taken for an example. That is, if the methylene blue of a constant rate is added in the test portion water solution of a constant rate and this is dropped at the matter measurement implement of this invention, extraction separation of the ion pair of a methylene blue and an anionic surfactant will be carried out into the organic phase of a matter measurement implement. The test portion water solution which adhered to the matter measurement implement using ion exchange water etc. after that is flushed, and whenever [coloring / of the organic phase which remained] is measured. Measurement of viewing or the amount of transmitted lights, and the amount of reflected lights can perform measurement of whenever [coloring]. Thus, if the matter measurement implement of this invention is used, special material can be measured by simple actuation.

[0043] Moreover, when using the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which comes to dissolve a nonaqueous solubility macromolecule in the ionophore list which carries out complexing to ion and shows a detectable response to the organic solvent of this invention, it can measure as follows.

[0044] It will be as follows if the case where bathocuproine is used as an ionophore for copper ions is taken for an example. The test portion water solution containing a copper ion is dropped at the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which makes it come to dissolve bathocuproine and a nonaqueous solubility macromolecule in an organic solvent. A test portion water solution permeates the whole ion measurement implement promptly, and a copper ion is extracted into an organic phase by the bathocuproine which exists in the organic phase of an ion measurement implement. Since an organic phase is three-dimension-network structure, thickness is thin dramatically, and this extract is performed promptly. Bathocuproine produces change in the absorption spectrum by complexing with the extracted copper ion. The

concentration of the copper ion contained in the test portion water solution can be known by flushing the test portion water solution which adhered to the ion measurement implement using ion exchange water etc. after fixed time amount progress, and measuring whenever [coloring / of the organic phase which remained]. Although it took the response several hours in the extraction method by the conventional film to be completed thoroughly, a response can be terminated within several minutes with the ion measurement implement of this invention. [0045] Furthermore, when it considers as the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which comes to dissolve the ionophore, the reporter coloring matter, and the nonaqueous solubility macromolecule which carry out complexing to ion in the organic solvent of this invention, it can measure as follows.

[0046] It will be as follows if the case where the potassium ion in a water solution is measured is taken for an example as an ionophore for potassiums, using a 2 and 6-dibromo-3'-methoxy indophenol as valinomycin and reporter coloring matter. A test portion water solution is dropped at the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which comes to dissolve a valinomycin, 2, and 6-dibromo-3'-methoxy indophenol and a nonaqueous solubility macromolecule in an organic solvent. It is necessary to keep pH of a test portion water solution constant using a buffer for pH etc. at this time. A test portion water solution permeates the whole ion measurement implement promptly, extraction separation of the potassium ion is carried out into an organic phase by the valinomycin which exists in the organic phase of an ion measurement implement, to it and coincidence, a 2 and 6-dibromo-3'-methoxy indophenol emits a hydrogen ion into a test portion water solution, and it colors. The potassium ion extracted in the film increases and the amount of the reporter coloring matter which colors in connection with it also increases it, so that there is much potassium ion in a test portion water solution. As a result, coloring of the measurement implement according to the amount of the potassium ion in a test portion water solution is observed, and the concentration of potassium ion can be known from whenever [coloring / of an ion measurement implement]. Since an organic phase is three-dimension-network structure, thickness is thin dramatically, this extract and bleedoff are performed promptly, and it has the features that the time amount which measurement takes is short.

[0047] Of course, the matter measurement implement of this invention is applicable to other well-known measurement principles which perform an extract or the ion exchange using the organic solvent of an organic solvent or nonaqueous solubility. It has the features that it can measure by the short time and simple actuation in any case.

[0048]

[Effect of the Invention] Since the matter measurement implement of this invention has the basic structure where the organic interphase of three-dimension-network structure which comes to dissolve a nonaqueous solubility macromolecule in a difficulty water solubility difficulty volatility organic solvent was filled up with the water soluble polymer phase, a test portion can permeate the whole matter measurement implement promptly, and since the thickness of an organic phase is small, it has the features that migration of the measuring object matter from a test portion to an organic phase is performed promptly. Therefore, it also has the features that it is simple actuation and extraction separation actuation which needed conventionally complicated actuation can be performed in a short time.

[0049] Therefore, a great profit is brought about in the blood intermediate ion density measurement in the case of high measurement of urgency, i.e., the site and operating room of emergency medical service. Moreover, the blood serum within the time amount restricted also in the general clinical laboratory test or the measurement size of a urine specimen is made to increase, and the increase in efficiency of an activity and carrying out smoothly of a diagnosis are brought about. Moreover, since it has complicated actuation, conventionally, measurement to which only the inside of a laboratory was carried out can also be performed in a test portion extraction site, and a measurement result can be known promptly.

[0050] Furthermore, since the conventional porosity support is not used for the matter

measurement implement of this invention, it can be manufactured at an easy process. Therefore, it has the features that it can manufacture cheaply. The above point, industrial worth of this invention is very large.

[0051]

[Example] Although an example and the example of a comparison are given and explained below in order to explain this invention still more concretely, this invention is not limited to these examples.

[0052] As a nonaqueous solubility giant molecule, 700mg of dioctyl sebacates was dissolved as 300mg (average degree of polymerization 1000) of polyvinyl chlorides, and an organic solvent, polyethylene-glycol (mean molecular weight 4000) 10g was dissolved in example 1 tetrahydrofuran 10ml as a water soluble polymer, and this was applied to it with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. When it is left in a room temperature and made to dry for 1 hour, the matter measurement implement which began to become cloudy gradually and obtained the white filmy material eventually and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the white filmy material fixed on the transparence polypropylene sheet was obtained.

[0053] The dipping of this matter measurement implement was carried out for 3 minutes into 100ml of ion exchange water, and the organic phase which remained on the transparence polypropylene sheet was observed with the electron microscope. In this case, the taken photograph is shown in drawing 1. It was checked that the network structure object of a three dimension is formed of the organic phase.

[0054] Example 2 1.5ml of 0.30M tetra-thio cyano cobalt (II) acid ammonium water solutions was added to 10ml of water solutions of the various concentration of the hepta-oxyethylene dodecylether which is the analysis nonionic surfactant of a nonionic surfactant, and it stirred for 1 minute. 20micro of this water solution was dropped at the matter measurement implement of an example 1 1 times, and about 2ml of ion exchange water washed the matter measurement implement after 1 minute. The result measured with the amount measuring device of reflected lights which shows the 330nm amount of reflected lights of a matter measurement implement in drawing 2 was shown in a table 1. It was shown that the quantum of a nonionic surfactant can be performed by simple actuation using the matter measurement implement of this invention.

[0055]

[A table 1]

表 1

ヘプタオキシエチレンドデシルエーテル濃度 (mg/100ml)	反射光量
0.1	0.97
0.6	0.95
0.9	0.93
1.5	0.91
2.0	0.89

Example 3 To ion measurement implement tetrahydrofuran 10ml for zinc ion As a nonaqueous solubility giant molecule, 300mg (average degree of polymerization 1000) of polyvinyl chlorides, As an organic solvent, as 700mg of dioctyl sebacates, and a water soluble polymer Polyvinyl-pyrrolidone 10g, 2-(5-BUROMO-2-pyridyl azo)-5-diethylamino phenol 7.3mg (21micromol) was dissolved as an ionophore for zinc ion, and this was applied with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. When it is left in a room temperature and made to dry for 1 hour, the ion measurement implement which began to become muddy gradually and obtained the dark reddish-brown filmy material eventually and

which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the filmy material fixed on the transparence polypropylene sheet was obtained. Thus, if ion exchange water washes the obtained ion measurement implement, it can obtain only the organic phase part colored dark reddish-brown. As the example 1 showed, this organic phase part had taken three-dimension-network structure.

[0056] Example 4 The amount measuring device of reflected lights which shows the ion measurement implement produced in the measurement example 3 of zinc ion in drawing 2 was equipped, and 25micro of water solutions 1 of various zinc chloride concentration was dropped. The amount of reflected lights with a wavelength of 553nm was measured, and the absorbance was calculated on the basis of the amount of reflected lights at the time of zinc chloride concentration 0mM. The result was shown in drawing 3. Moreover, the time amount taken to become the fixed amount of reflected lights was also measured. Each required time amount was within the limits of 15 - 20 seconds.

[0057] 2-(5-BUROMO-2-pyridyl azo)-5-diethylamino phenol 7.3mg (21micromol) was dissolved in example of comparison 1 tetrahydrofuran 10ml as an ionophore 700mg of dioctyl sebacates, and for zinc ion as a nonaqueous solubility giant molecule as 300mg (average degree of polymerization 1000) of polyvinyl chlorides, and an organic solvent, and this was applied to it with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. The measurement implement which left it in the room temperature, was dried for 1 hour, and obtained the filmy material of transparence dark reddish-brown and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the filmy material fixed on the transparence polypropylene sheet was obtained.

[0058] By the same actuation as an example 4, various zinc chloride concentration water solutions were measured for this measurement implement. However, since the fixed amount of reflected lights was not reached even if 5 minutes passed after the water solution was dropped, it was not able to measure.

[0059] It dried, after ion exchange water washed the measurement implement produced in the example of comparison 2 example 3, and the water soluble polymer was removed, and the measurement implement which comes to dissolve a nonaqueous solubility macromolecule in the organic solvent of three-dimension-network structure was produced. By the same actuation as an example 4, various zinc chloride concentration water solutions were measured for this measurement implement. However, even after the water solution was dropped, the water solution did not permeate into the micropore of a measurement implement, and most change of a color was not seen. Since the fixed amount of reflected lights was not reached even if 5 minutes passed, it was not able to measure.

[0060] It was shown from examples 3 and 4, the example 1 of a comparison, and the example 2 of a comparison that it is necessary to fill up with the water soluble polymer for performing measurement in a short time at the organic interphase of three-dimension-network structure.

[0061] The tetrahydrofuran solution was applied on the transparence polypropylene sheet by the same presentation and the actuation as example of comparison 3 example 3, and it was made to dry quickly using a blower. The solution became cloudy rapidly. The measurement implement which was dried after that for 1 hour and obtained the filmy material of opaque dark reddish-brown and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the massive object fixed on the transparence polypropylene sheet was obtained.

[0062] By the same actuation as an example 4, various zinc chloride concentration water solutions were measured for this measurement implement. The result was shown in drawing 4. Since the response obtained since the part to which an about 0.1mm hole does not penetrate an aperture and light does not penetrate the film partly to a measurement implement by dropping of a water solution is made was very small, the unsuitable thing was shown in measurement.

[0063] Examples 3 and 4 and example 3 of a comparison, It was shown that it is the making [deposit / the deposit of the organic phase which becomes a difficulty water solubility difficulty volatility organic solvent list from a nonaqueous solubility macromolecule at least performs deposit actuation to the bottom of the condition which starts before a deposit of a water soluble polymer, and / fill up into the organic interphase of three-dimension-network structure with the

manufacture of the matter measurement implement of this invention by which the laminating was carried out on the base material]-water soluble polymer phase need.

[0064] Example 5 To ion measurement implement chloroform 10ml for sodium ion As a nonaqueous solubility giant molecule, as polystyrene 300mg and an organic solvent o-nitrophenyl octyl ether 600mg, As a water soluble polymer, polyethylene-glycol (average molecular weight 6000) 10g, They are 5, 11, 17, and 23-tetra-t-butyl as a sodium ionophore. - 25, 26, 27, 28 - Tetrakis (ethoxycarbonyl)-methoxy-calyx [4] allene 18.4mg (18.4micromol), 2 and 6-dibromo-3'-methoxy indophenol 7.1mg (18.4micromol) was dissolved as coloring matter, and this was applied with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. When it was made to leave and dry in a room temperature, the ion measurement implement which turbidity went up gradually and obtained the filmy material of a brown white color eventually and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the filmy material of a brown white color fixed on the transparence polypropylene sheet was obtained.

[0065] The amount measuring device of reflected lights which shows the ion measurement implement produced in the example 6 example 5 in drawing 2 was equipped, and 25micro (it prepares with a potassium hydroxide to pH5.5) of 2-morpholino ethane-sulfonic-acid 10mM buffer solutions I of various sodium chloride concentration was dropped. The amount of reflected lights with a wavelength of 670nm was measured, and the absorbance was calculated on the basis of the amount of reflected lights at the time of sodium chloride concentration 0mM. The result was shown in drawing 5. Drawing 5 shows that sodium ion can be measured between 10-0.1mM(s). Moreover, the time amount taken to become the fixed amount of reflected lights was also measured. Each required time amount was within the limits of 15 - 20 seconds.

[0066] To example of comparison 4 tetrahydrofuran 10ml, as a nonaqueous solubility giant molecule, 300mg (average degree of polymerization 1000) of polyvinyl chlorides, As an organic solvent, o-nitrophenyl octyl ether 600mg, They are 5, 11, 17, and 23-tetra-t-butyl as a sodium ionophore. - 25, 26, 27, 28 - Tetrakis (ethoxycarbonyl)-methoxy-calyx [4] allene 18.4mg (18.4micromol), 2 and 6-dibromo-3'-methoxy indophenol 7.1mg (18.4micromol) was dissolved as coloring matter, and this was applied with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. It was made to leave and dry in a room temperature, and the measurement implement was obtained as a filmy material of uniform transparence brown.

[0067] Thus, the amount measuring device of reflected lights which shows the manufactured measurement implement in drawing 2 was equipped, and 25micro (it prepares with a potassium hydroxide to pH5.5) of 2-morpholino ethane-sulfonic-acid 10mM buffer solutions I of various sodium chloride concentration was dropped. Although the amount of reflected lights with a wavelength of 670nm was measured, even if 3 minutes passed, the fixed amount of reflected lights was not reached. It was not able to measure on the same conditions as an example 6 that a speed of response is slow.

[0068] It was shown that it can measure promptly using the ion measurement implement of this invention from an example 6 and the example 5 of a comparison.

[0069] The ion measurement implement for sodium ion was produced by the same actuation as an example 5 using the nonaqueous solubility macromolecule shown in seven to example 5-10 of comparison, and example 227 table 2, an organic solvent, a water soluble polymer, the same sodium ionophore as an example 5, and coloring matter. The amount measuring device of reflected lights which shows the produced measurement implement in drawing 2 was equipped, and 25micro (it prepares with a potassium hydroxide to pH5.5) of sodium chloride concentration 10 2-morpholino ethane-sulfonic-acid 10mM buffer solutions I of mM was dropped. The amount of reflected lights with a wavelength of 670nm was measured, and time amount until the amount of reflected lights at that time becomes fixed was measured. The result was shown in a table 2. The quick response was not obtained, even if missing in the nonaqueous solubility macromolecule, the organic solvent, or the water soluble polymer so that examples 5-10 might show. Moreover, it was shown that a quick response is obtained in the combination of various nonaqueous solubility macromolecules, an organic solvent, and a water soluble polymer from examples 7-22.

[0070]

[A table 2]

表 2

	非水溶性高分子	有機溶剤	水溶性高分子	応答時間 (秒)
比較例 5	なし	セバシン酸ジオクチル	ポリエチレングリコール	180 秒以上
比較例 6	〃	フタル酸ジオクチル	ポリエチレングリコール	180 秒以上
比較例 7	〃	〃	ポリビニルピロリドン	180 秒以上
比較例 8	〃	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	180 秒以上
比較例 9	ポリ塩化ビニル	なし	ポリエチレングリコール	反射量変化なし
比較例 10	〃	セバシン酸ジオクチル	なし	180 秒以上
実施例 7	〃	〃	ポリビニルピロリドン	17
実施例 8	〃	〃	ポリエチレングリコール	18
実施例 9	〃	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 10	〃	〃	ポリビニルピロリドン	20
実施例 11	〃	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	18
実施例 12	〃	〃	ポリビニルピロリドン	18
実施例 13	ポリメタクリル酸メチル	セバシン酸ジオクチル	ポリエチレングリコール	18
実施例 14	〃	〃	ポリビニルピロリドン	18

[0071]

[A table 3]

表 2 (続き)

	非水溶性高分子		水溶性高分子	応答時間 (秒)
実施例 1 5	ポリメタクリル酸メチル	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 1 6	"	"	ポリビニルピロリドン	20
実施例 1 7	"	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	17
実施例 1 8	ポリスチレン	セバシン酸ジオクチル	ポリエチレングリコール	18
実施例 1 9	"	"	ポリビニルピロリドン	19
実施例 2 0	"	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 2 1	"	"	ポリビニルピロリドン	19
実施例 2 2	"	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	18

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the ion measurement implement which used this for the matter measurement implement list which can be used for detection of various matter in a solution, and a quantum.

[0002]

[Description of the Prior Art] In the field of analytical chemistry, measuring various matter especially in a water solution, and using for medical care, a production activity, environmental protection, etc. is performed among the solution. For example, it is known by measuring the ion concentration in the blood serum of the body, and urine that the useful information about a patient's symptoms will be obtained. Moreover, measurement of the concentration of the surfactant contained in the monitor of the harmful heavy metal in the waste fluid discharged as a result of the production activity of an enterprise, domestic wasted water, etc. is needed for environmental protection. In addition, although there will be no enumeration if an example is given, measurement of the matter in a solution is not dispensable to our life.

[0003] The measuring method of the matter in a solution changes with the measuring object matter, and many approaches exist. There is the approach of calling a colorimetric method which measures the absorption by which induction is carried out with the measuring object matter into it, or change of a fluorescence spectrum. In a colorimetric method, extraction separation of the measuring object matter in a water solution is carried out into an organic solvent, and a quantum is performed in many cases from the variation of the absorption produced by the interaction of the measuring object matter which moved to the organic solvent, and the color reagent beforehand added underwater or in the organic solvent, or a fluorescence spectrum.

[0004] Extraction separation is actuation to which a certain matter is made to separate into another side from one side of two sorts of liquids and which is performed for accumulating. Actuation of taking out only the near solution which two sorts of solutions which generally are not mixed mutually were put into the separating funnel, this was shaken, and the measuring object matter moved after that is performed. Therefore, in measurement of the colorimetric method for which extraction separation actuation is needed, the tool of dedication for extraction separation is needed. In order to compensate this fault, using the film which consists of a difficulty water solubility difficulty volatility organic solvent which added the nonaqueous solubility macromolecule instead of the organic solvent is known. In volume [40th] (1991) 227-231 pages for example, "analytical chemistry" — The film made to dissolve the bathocuproine which forms a complex specifically with the copper ion of monovalence, and is colored in a nonaqueous solubility macromolecule and a difficulty water solubility difficulty volatility organic solvent is used. Carry out extraction separation of the copper ion into the film, the bathocuproine in the film is made to color, and the approach of measuring copper ion concentration from a membranous absorbance is described by by contacting this film in the water solution containing the copper ion of the monovalence of strange concentration. However, since the mass transfer that the measuring object matter in a water solution moves into a difficulty water solubility difficulty volatility organic solvent in extraction separation actuation

was indispensable, diffusion in the difficulty water solubility difficulty volatility organic solvent of the measuring object matter became rate-limiting, and there was demerit in which the time amount concerning measurement was very long.

[0005] The analysis method of the ion by the colorimetric method of the principle which produces absorption or change of a fluorescence spectrum by migration of the measuring object matter is stated to JP,59-211864,A, JP,59-231452,A, and JP,60-194360,A. These are ion concentration measurement implements which consist of a difficulty water solubility difficulty volatility organic solvent containing various ionophores and the coloring matter called the reporter matter. In these measurement implements, by making the test portion water solution containing the ion which is the measuring object matter contact, migration into the film of ion breaks out and change is produced in the extinction spectrum or fluorescence spectrum of coloring matter. It is known that the rate which ion diffuses within the film also in this approach will become rate-limiting. It is known that it is effective technique to make thickness thin to about several micrometers. However, while it becomes large, since the rate of coloring change has the fault that the coloring variation obtained becomes small, it is not practical.

[0006] Various technique has been developed in order to conquer the above-mentioned fault. For example, the ion concentration measurement implement which made porosity support support the difficulty water solubility difficulty volatility organic solvent containing pH susceptibility coloring matter called various ionophores and the reporter matter to JP,60-194360,A is indicated. It was the attempt in which both the rate of coloring change and the problem of coloring variation would be solved, by making the front face of porosity support like a filter paper cover thinly an ionophore and the difficulty water solubility difficulty volatility organic solvent containing the reporter matter. A certain amount of solution was able to be carried out by this technique. However, when the measurement implement using these porosity support manufactured, it needed the process of fixing porosity support on a base material in an exact location, and in addition, manufacturing to a large quantity and homogeneity had difficulty or the fault that cost started dramatically. Therefore, development of technique with the effectiveness same [without using porosity support] was desired.

[0007]

[Problem(s) to be Solved by the Invention] When it is used for the matter measurement implement which can perform measurement which combines practically sufficient speed of response and the amount of responses, for example, a colorimetric method, it aims at development of the matter measurement implement which can perform measurement which combines practically sufficient coloring rate and coloring variation, and the ion measurement implement which used this for the list.

[0008]

[A means to solve a technical problem] this invention persons repeated research wholeheartedly that the matter measurement implement which can solve this technical problem should be developed. Consequently, when the organic phase and water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule in a difficulty water solubility difficulty volatility organic solvent took specific structure, it found out demonstrating sufficient engine performance solving the above-mentioned technical problem.

[0009] That is, this invention is (A). It consists of the organic phase and (B) water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule in a difficulty water solubility difficulty volatility organic solvent, (A) organic phase is the matter measurement implement characterized by (B) water soluble polymer phase being filled up with three-dimension-network structure by the nothing (** A) organic interphase, and it is the matter measurement implement with which it comes to carry out the laminating of this matter measurement implement on a base material further.

[0010] Other invention is (A). Coloring matter which carries out a mutual reaction and shows a detectable response to the ionophore and this complexing which carry out complexing to the ionophore or ion which carries out complexing to ion and shows a detectable response to a difficulty water solubility difficulty volatility organic solvent, It consists of the organic phase and (B) water soluble polymer phase which come to dissolve a nonaqueous solubility macromolecule

in a list. (A) An organic phase is the ion measurement implement characterized by (B) water soluble polymer phase being filled up with three-dimension-network structure by the nothing (** A) organic interphase, and it is the ion measurement implement with which it comes to carry out the laminating of this ion measurement implement on a base material further.

[0011] Furthermore, other invention, It is the manufacture approach of the above-mentioned matter measurement implement characterized by evaporating an organic solvent under the condition which the solution which comes to dissolve a difficulty water solubility difficulty volatility organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer in an organic solvent is applied on a base material, and the deposit of the organic phase which subsequently consists of a difficulty water solubility difficulty volatility organic solvent and a nonaqueous solubility macromolecule starts before a deposit of a water soluble polymer, or deposits in coincidence.

[0012] The organic phase which constitutes the matter measurement implement of this invention consists of a difficulty water solubility difficulty volatility organic solvent and a nonaqueous solubility macromolecule fundamentally.

[0013] The above-mentioned difficulty water solubility difficulty volatility organic solvent (only henceforth an organic solvent) points out the thing of an organic compound liquefied in the ordinary temperature which does not have the functional group which is a high-boiling point and can be ionized refractory to water. This organic solvent is used in order to extract the measuring object matter in a test portion and to dissolve coloring matter required for measurement etc. The boiling point is specifically 100-degree more than Centigrade, and it is desirable in order to carry out measurement by which it was stabilized that the organic solvent which can dissolve to 1l. of water is 1g or less extent.

[0014] If the organic solvent used suitably is mentioned concretely, there is high-boiling point ether, such as high-boiling point ester, such as adipic-acid dibutyl and a dioctyl phthalate, o-nitrophenyl octyl ether, and diphenyl ether.

[0015] As a nonaqueous solubility macromolecule, it can dissolve in the above-mentioned organic solvent, and an insoluble macromolecule is used for water. This nonaqueous solubility macromolecule dissolves in the above-mentioned organic solvent, and gives the viscosity and physical reinforcement suitable for the activity as a matter measurement implement.

[0016] If the example of the nonaqueous solubility giant molecule used suitably is given, there are styrene system polymers, such as methacrylic ester system polymers, such as vinylidene-chloride system polymers, such as polyvinyl chloride system polymers, such as a polyvinyl chloride and an ethylene-vinyl chloride copolymer, a vinyl chloride-vinylidene-chloride copolymer, and a vinylidene chloride, and a polymethyl methacrylate, and polystyrene, etc. Although it can be used without being limited especially about the polymerization degree of a nonaqueous solubility macromolecule, if a with an average degree of polymerization of 500 or more thing is generally used, an organic solvent comes to show free-standing, and the handling when considering as a matter measurement implement it being the range where it is below the upper limit of the solubility, and an organic solvent shows free-standing becomes easy and is suitable for the concentration in the organic solvent of a desirable nonaqueous solubility macromolecule. Although the density range changes with classes of the nonaqueous solubility macromolecule to be used and organic solvent, it is suitable to make it dissolve five to 100% of the weight, and to use a nonaqueous solubility macromolecule to an organic solvent, generally. Especially the dissolution approach to the organic solvent of a nonaqueous solubility macromolecule is not limited, but it is dissolved, usually adding a nonaqueous solubility macromolecule gradually into an organic solvent.

[0017] The water soluble polymer phase which constitutes this invention consists of a water soluble polymer fundamentally. This water soluble polymer is used in order to promote that a test portion permeates between the organic phases of three-dimension-network structure promptly.

[0018] A water soluble polymer is a macromolecule which can dissolve in water, and the solubility is [a water soluble polymer] 50 wt%. It is desirable to use the above thing. If a water soluble polymer does not exist, since can go a test portion and it cannot be crossed even to all

the corners of an organic solvent, the measuring time required when a matter measurement implement is used for measurement will become long. It is desirable to use a thing refractory to an organic solvent for a water soluble polymer. Even if it uses what is dissolved in an organic solvent, the effectiveness of this invention can be acquired, but when contacting the matter measurement implement of a test portion and this invention in that case, stable measurement may not be able to be performed with remarkable deformation. Moreover, when the ease of the handling at the time of using it as a matter measurement implement is taken into consideration, it is desirable to use the thing of the polymerization degree which will be in a solid state in a room temperature. Maintaining suitable physical reinforcement to the amount of whole including an organic solvent and a nonaqueous solubility macromolecule, if the amount of a water soluble polymer is 50 – 100% of the weight of the range, although osmosis of a test portion is promoted, it is desirable.

[0019] If the example of the water soluble polymer which can be used suitably is given, there are polyether system high molecular compounds, such as a polyethylene glycol and a polypropylene glycol, a polyvinyl pyrrolidone, polyvinyl alcohol, polyacrylamide, etc.

[0020] It can check that the organic phase of this invention is three-dimension-network structure by observing the part which remains after flushing a water soluble polymer with water with an electron microscope etc.

[0021] Although various approaches can be used for the approach of making a water soluble polymer phase existing in the organic interphase of three-dimension-network structure in the state of restoration, the approach used most suitably is as follows.

[0022] That is, it is made to dissolve in an organic solvent, and all of an organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer are applied on a base material after that, and they are dried. The micro phase separation of an organic phase and a water soluble polymer phase which comes to dissolve a nonaqueous solubility macromolecule in an organic solvent occurs with evaporation of an organic solvent, and where the organic phase of three-dimension-network structure is filled up with a water soluble polymer phase, it deposits.

[0023] If all of an organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer are dissolved as an organic solvent used for the above-mentioned approach, it can use that there is no limit in any way. Although the organic solvents which can be used with the combination of the selected organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer differ, if the example is given, there are alkyl halides, such as ether, such as a tetrahydrofuran, dichloromethane, chloroform, and a dichloroethane. Since the amount of the organic solvent used changes with solubility of the selected organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer, it cannot generally specify, but in order to urge a deposit, it is common to carry out an organic solvent, a nonaqueous solubility macromolecule, or a water soluble polymer to an organic solvent and more than tales doses.

[0024] By the above-mentioned depositing method, when the deposit of the organic phase which consists of an organic solvent and a nonaqueous solubility macromolecule starts before the deposit of a water soluble polymer phase or it deposits in coincidence, uniform and good structure can be acquired. The case where it manufactures on the conditions from which the deposit of an organic phase begins previously is more suitable. On the contrary, when a deposit of a water soluble polymer starts before the deposit of an organic phase, the matter measurement implement obtained becomes an ununiformity and what has many openings, and it is not suitable for measurement by the colorimetric method.

[0025] Control of the deposit sequence of an organic phase and a water soluble polymer phase can be carried out by controlling the solubility over the organic phase and water soluble polymer phase of the organic solvent in which a bi-phase is dissolved. Generally this soluble control is controlled by the polarity of an organic solvent. Being easy to dissolve the organic solvent and nonaqueous solubility macromolecule which generally constitute an organic phase in a polar low organic solvent, a water soluble polymer has conversely the property to be easy to dissolve in a polar high organic solvent. Therefore, if a polar high organic solvent is used, the deposit of an organic phase with low solubility will start previously. Therefore, the deposit of the organic phase which consists of an organic solvent and a nonaqueous solubility macromolecule can be started

before the deposit of a water soluble polymer phase.

[0026] The polarity of an organic solvent is controllable by mixing two or more sorts of organic solvents or water. If the example used suitably is shown, there are a tetrahydrofuran which added about 0.1% of water, chloroform which added the methanol 1%. Moreover, also when an ionophore and coloring matter are dissolved in an organic phase, the sequence of a deposit can be controlled by the same approach of being satisfactory at all.

[0027] If the concrete example which obtains the matter measurement implement of good structure is given, as an organic solvent, a polymethyl methacrylate can be used as o-nitrophenyl octyl ether and a nonaqueous solubility giant molecule, a polyethylene glycol (average molecular weight 6000 [about]) can be used as a water soluble polymer, these can be dissolved in a tetrahydrofuran, and it can apply on a glass plate, and can produce by making it leave and dry in a room temperature. If the moisture of a minute amount exists in a tetrahydrofuran at this time, a deposit of a polyethylene glycol becomes slow and can acquire the above-mentioned structure with sufficient repeatability. By using the organic phase in which the organic colored phase or coloring matter was dissolved, when the colored sludge deposits, it can know that the deposit of the organic phase which consists of an organic solvent and a nonaqueous solubility macromolecule will start before the deposit of a water soluble polymer phase.

[0028] the principle and approach of using for measurement of ion concentration the organic phase which comes to dissolve the ionophore which carries out complexing to ion and shows a detectable response to an organic solvent -- well-known -- "analytical chemistry" -- volume [40th] (1991) 227-231 pages have the description. Moreover, the principle and approach of using for measurement of ion concentration the coloring matter which carries out a mutual reaction and shows a detectable response to the ionophore and this complexing which carry out complexing to ion at an organic solvent, and the organic phase which comes to dissolve a nonaqueous solubility macromolecule in a list are also well-known, and JP,59-211864,A, "analytical Science" (ANALYTICAL SCIENCE) 1990 year 715-720 page, etc. have the description. The ionophore and coloring matter which are used in this invention can use satisfactory at all what is stated by these reference.

[0029] In this invention, an ionophore is a compound with the property which carries out a specific cation and specific complexing. The coloring matter which has the capacity which forms a specific metal ion and a specific complex, produces change in the electron orbit condition by forming this metal ion and a complex, and produces change in the absorption spectrum or fluorescence spectrum of a complex can be used as an ionophore which carries out complexing to the ion of this invention, and shows a detectable response. As such an ionophore, there are nitroso aminophenols, diethylamino phenols, porphyrins, and phenoxazine.

[0030] Moreover, as an ionophore which can be used in the combination of the coloring matter which carries out a mutual reaction and shows a detectable response to the ionophore which carries out complexing to the ion of this invention, and the complexing concerned, there are several sorts of well-known cyclic peptide compounds, annular or an un-annular polyether derivative, a POTANDO derivative, a calyx allene derivative, etc.

[0031] If the example which can be used suitably [the ionophore which forms ion and a complex and shows a detectable response] is given potassium ion -- receiving -- 3 -- '- nitro-4' - (2, 4, 6-trinitro phenylamino) benzo-18-crown -6 and 4' - (2, 6-dinitro-4-trifluoro methylphenyl) amino benzo-15-crown-5 grade To a copper ion, bathocuproine etc. receives iron ion. Nitroso aminophenols, such as 2-nitroso-5-N and N-dipropylamino phenol As opposed to nickel and zinc ion diethylamino phenols, such as a 2-(5-BUROMO-2-pyridyl azo)-5-diethylamino phenol To a hydrogen ion, there are azo phenols, such as phenoxazine, such as [9-(diethylamino)-5-octadecanoyl imino-5H-[benzoa] phenoxazine], and a [5-OKUTADEKANO yloxy-2-(4-nitro phenylazo) phenol].

[0032] If the example which can be used suitably [the ionophore used for the ionophore which carries out complexing to ion and the complexing concerned on the other hand in the combination of the coloring matter in which a mutual reaction is carried out and a detectable response is shown] is given As a lithium ionophore, it is 6 and 6-dibenzyl. - 1, 4, 8, 11-tetra-OKISA cyclo tetradecane, 2, the 9-dibutyl -1, 10-phenanthroline, etc. are 5, 11, 17, and 23-

tetra-*t*-butyl as a; sodium ionophore. - It is 25, 26, 27, and 28. - Tetrakis (ethoxycarbonyl)-methoxy-calyx [4] allene, Screw [(12-crown -4) methyl] methyl dodecyl malonate etc. as a; potassium ionophore Valinomycin, 2, 3-naphth - 1, 4, 7, 10, 13-PENTAOKISA cyclo PENTADEKA-2-en, Screw [(benzo-15-crown -5) -4'-methyl] PIMERETO, Valinomycin etc. as a; calcium ionophore ** - diethyl - N - N - ' - [(4R, 5R) - four - five - dimethyl - one - eight - dioxo - three - six - dioxa - octamethylene -] - a screw (12-methyl friend throat decanoate) - etc. - as; magnesium ionophore - The benzo-15-thia crown-4-ether etc. has ** N, N'-diheptyl-N, an N'-dimethyl ASUPARUTO amide, etc. to a copper ion.

[0033] Of course, in addition to this, the compound with which forming a cation and a complex is known exists, and can also use them. As reference about an ionophore, the volume Yoshihisa Inoue and on George W.Gokel "Cation Binding by Macrocycles" (MARCEL DEKKER INCORPORATION 1990), 2013 - 2016 pages (ANALYTICAL CHEMISTRY) (1988) of work "application to analytical chemistry of functional macrocyclic compound" (IPC, Inc. 1990 issuance) 60-volume Analytical Chemistry edited by Hiroyuki Takeda, "Analytical chemistry" 23 pages [1412 - 1430 etc.] (1974) etc. are mentioned.

[0034] There is a series of coloring matter called a pH indicator or pH susceptibility coloring matter as coloring matter (it is henceforth described as reporter coloring matter) which carries out a mutual reaction and shows a detectable response to the ionophore and this complexing which carry out complexing to above-mentioned ion.

[0035] Since the reporter coloring matter used in this invention is solubilized by the organic solvent and contained, what has it is used. [remarkable water solubility and small] The reporter coloring matter of 5-10 is suitably used for electric dissociation exponent from extent of the coloring response when using it as the ease of the acquisition, and an ion measurement implement.

[0036] If reporter coloring matter is shown concretely, there are triphenylmethane dye derivatives, such as indophenol derivatives, such as a phenolphthalein derivative [, such as a phenolphthalein and a tetrabromo phenolphthalein,], 2, 6-dichloroindophenol, 2, and 6-dibromo-3'-methyl indophenol, and Victoria pure blue, etc.

[0037] An ionophore and reporter coloring matter can be dissolved in an organic solvent to the upper limit of the solubility. At this time, it is known that it is more suitable for the mole ratio of an ionophore and reporter coloring matter than that principle that it is 1:1. Generally, the concentration in the inside of the organic solvent of an ionophore and reporter coloring matter takes the sensibility when measuring into consideration, and it is desirable that it is the range of 0.1-200mM, respectively.

[0038] In this invention, it points out that the organic phase has network structure in three dimension like sponge and sponge as the organic phase which comes to dissolve a nonaqueous solubility giant molecule in an organic solvent is three-dimension-network structure. As for the matter with the same structure, various things are used with different construction material as the absorptivity matter or an object for filtration separation. The process of the matter which consists of a nonaqueous solubility macromolecule with three-dimension-mesh-like structure is indicated by JP,2-302449,A, JP,58-104940,A, JP,47-4593,A, etc. The matter of such conventional three-dimension-network structures makes a nonaqueous solubility macromolecule a subject, in order to realize high reinforcement, and generally it does not add an organic solvent. Moreover, even if the organic solvent is added, what was filled up with the water soluble polymer is not known. The organic phase which comes to dissolve a nonaqueous solubility macromolecule in the organic solvent of the three-dimension-network structure of this invention makes a subject the organic solvent for performing extract operation, and has a fundamental difference that a nonaqueous solubility macromolecule is added in order to give moderate viscosity and physical reinforcement.

[0039] In this invention, a test portion permeates in the micropore of the organic phase of three-dimension-network structure promptly by filling up the organic interphase of three-dimension-network structure with the water soluble polymer phase. since [moreover,] an organic phase is three-dimension-network structure -- several micrometers or less -- thickness can be carried out, consequently migration into the organic phase of the measuring object matter can be

completed promptly. Moreover, the aperture of the micropore made when the water soluble polymer of the organic phase of three-dimension-network structure is removed can take uniform structure, when it is 500 micrometers or less. By taking uniform structure, more exact measurement is enabled and it is desirable.

[0040] Although the matter measurement implement of this invention can be used for measurement even if it remains as it is, it can take into consideration an activity in combination with reagents, such as operability in the case of measurement, and a buffer for pH, etc., and it can carry out a laminating on a base material. A well-known approach can be used without limiting especially as the approach of a laminating. How to produce the matter measurement implement of this invention beforehand, and paste up with suitable adhesives, a double-sided tape, etc., if the example used suitably is given. The solution which comes to dissolve an organic solvent, a nonaqueous solubility macromolecule, and a water soluble polymer in an organic solvent can be applied on a base material, and the organic phase which comes to dissolve a nonaqueous solubility macromolecule in an organic solvent as above-mentioned can measure concentration of various matter using the matter measurement implement with the approach of drying under the conditions which deposit in three-dimension-network structure etc. which carried out in this way and was produced. It explains to below with an example.

[0041] Measurement of the concentration of the surfactant of the surfactant in current and a water solution adds specifically association or the coloring matter which carries out complexing in a water solution with a surfactant, extracts both a surfactant and the coloring matter to the extract of nonaqueous solubility out of a water solution, and is performed by measuring whenever [coloring / of an extract] after that. Such an approach is shown in Japanese Industrial Standards (JIS K 0102-1986 testing method for industrial waste water), and the methylene-blue absorptiometry, the tetra-thio cyano cobalt (II) acid absorptiometry, etc. are indicated. After each of these approaches puts two kinds of liquid into a container, in order to accompany them by actuation of shaking and extracting, they has the fault of requiring serious time amount and a serious effort.

[0042] The above-mentioned measurement can be performed by the same principle using the matter measurement implement of this invention. It will be as follows if a methylene-blue absorptiometry is taken for an example. That is, if the methylene blue of a constant rate is added in the test portion water solution of a constant rate and this is dropped at the matter measurement implement of this invention, extraction separation of the ion pair of a methylene blue and an anionic surfactant will be carried out into the organic phase of a matter measurement implement. The test portion water solution which adhered to the matter measurement implement using ion exchange water etc. after that is flushed, and whenever [coloring / of the organic phase which remained] is measured. Measurement of viewing or the amount of transmitted lights, and the amount of reflected lights can perform measurement of whenever [coloring]. Thus, if the matter measurement implement of this invention is used, special material can be measured by simple actuation.

[0043] Moreover, when using the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which comes to dissolve a nonaqueous solubility macromolecule in the ionophore list which carries out complexing to ion and shows a detectable response to the organic solvent of this invention, it can measure as follows.

[0044] It will be as follows if the case where bathocuproine is used as an ionophore for copper ions is taken for an example. The test portion water solution containing a copper ion is dropped at the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which makes it come to dissolve bathocuproine and a nonaqueous solubility macromolecule in an organic solvent. A test portion water solution permeates the whole ion measurement implement promptly, and a copper ion is extracted into an organic phase by the bathocuproine which exists in the organic phase of an ion measurement implement. Since an organic phase is three-dimension-network structure, thickness is thin dramatically, and this extract is performed promptly. Bathocuproine produces change in the absorption spectrum by complexing with the extracted copper ion. The

concentration of the copper ion contained in the test portion water solution can be known by flushing the test portion water solution which adhered to the ion measurement implement using ion exchange water etc. after fixed time amount progress, and measuring whenever [coloring / of the organic phase which remained]. Although it took the response several hours in the extraction method by the conventional film to be completed thoroughly, a response can be terminated within several minutes with the ion measurement implement of this invention. [0045] Furthermore, when it considers as the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which comes to dissolve the ionophore, the reporter coloring matter, and the nonaqueous solubility macromolecule which carry out complexing to ion in the organic solvent of this invention, it can measure as follows.

[0046] It will be as follows if the case where the potassium ion in a water solution is measured is taken for an example as an ionophore for potassiums, using a 2 and 6-dibromo-3'-methoxy indophenol as valinomycin and reporter coloring matter. A test portion water solution is dropped at the ion measurement implement with which it comes to fill [a water soluble polymer phase] up the organic interphase of three-dimension-network structure which comes to dissolve a valinomycin, 2, and 6-dibromo-3'-methoxy indophenol and a nonaqueous solubility macromolecule in an organic solvent. It is necessary to keep pH of a test portion water solution constant using a buffer for pH etc. at this time. A test portion water solution permeates the whole ion measurement implement promptly, extraction separation of the potassium ion is carried out into an organic phase by the valinomycin which exists in the organic phase of an ion measurement implement, to it and coincidence, a 2 and 6-dibromo-3'-methoxy indophenol emits a hydrogen ion into a test portion water solution, and it colors. The potassium ion extracted in the film increases and the amount of the reporter coloring matter which colors in connection with it also increases it, so that there is much potassium ion in a test portion water solution. As a result, coloring of the measurement implement according to the amount of the potassium ion in a test portion water solution is observed, and the concentration of potassium ion can be known from whenever [coloring / of an ion measurement implement]. Since an organic phase is three-dimension-network structure, thickness is thin dramatically, this extract and bleedoff are performed promptly, and it has the features that the time amount which measurement takes is short.

[0047] Of course, the matter measurement implement of this invention is applicable to other well-known measurement principles which perform an extract or the ion exchange using the organic solvent of an organic solvent or nonaqueous solubility. It has the features that it can measure by the short time and simple actuation in any case.

[0048]

[Effect of the Invention] Since the matter measurement implement of this invention has the basic structure where the organic interphase of three-dimension-network structure which comes to dissolve a nonaqueous solubility macromolecule in a difficulty water solubility difficulty volatility organic solvent was filled up with the water soluble polymer phase, a test portion can permeate the whole matter measurement implement promptly, and since the thickness of an organic phase is small, it has the features that migration of the measuring object matter from a test portion to an organic phase is performed promptly. Therefore, it also has the features that it is simple actuation and extraction separation actuation which needed conventionally complicated actuation can be performed in a short time.

[0049] Therefore, a great profit is brought about in the blood intermediate ion density measurement in the case of high measurement of urgency, i.e., the site and operating room of emergency medical service. Moreover, the blood serum within the time amount restricted also in the general clinical laboratory test or the measurement size of a urine specimen is made to increase, and the increase in efficiency of an activity and carrying out smoothly of a diagnosis are brought about. Moreover, since it has complicated actuation, conventionally, measurement to which only the inside of a laboratory was carried out can also be performed in a test portion extraction site, and a measurement result can be known promptly.

[0050] Furthermore, since the conventional porosity support is not used for the matter

measurement implement of this invention, it can be manufactured at an easy process. Therefore, it has the features that it can manufacture cheaply. The above point, industrial worth of this invention is very large.

[0051]

[Example] Although an example and the example of a comparison are given and explained below in order to explain this invention still more concretely, this invention is not limited to these examples.

[0052] As a nonaqueous solubility giant molecule, 700mg of dioctyl sebacates was dissolved as 300mg (average degree of polymerization 1000) of polyvinyl chlorides, and an organic solvent, polyethylene-glycol (mean molecular weight 4000) 10g was dissolved in example 1 tetrahydrofuran 10ml as a water soluble polymer, and this was applied to it with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. When it is left in a room temperature and made to dry for 1 hour, the matter measurement implement which began to become cloudy gradually and obtained the white filmy material eventually and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the white filmy material fixed on the transparence polypropylene sheet was obtained.

[0053] The dipping of this matter measurement implement was carried out for 3 minutes into 100ml of ion exchange water, and the organic phase which remained on the transparence polypropylene sheet was observed with the electron microscope. In this case, the taken photograph is shown in drawing 1. It was checked that the network structure object of a three dimension is formed of the organic phase.

[0054] Example 2 1.5ml of 0.30M tetra-thio cyano cobalt (II) acid ammonium water solutions was added to 10ml of water solutions of the various concentration of the hepta-oxyethylene dodecylether which is the analysis nonionic surfactant of a nonionic surfactant, and it stirred for 1 minute. 20micro of this water solution was dropped at the matter measurement implement of an example 1 1 times, and about 2ml of ion exchange water washed the matter measurement implement after 1 minute. The result measured with the amount measuring device of reflected lights which shows the 330nm amount of reflected lights of a matter measurement implement in drawing 2 was shown in a table 1. It was shown that the quantum of a nonionic surfactant can be performed by simple actuation using the matter measurement implement of this invention.

[0055]

[A table 1]

表 1

ヘプタオキシエチレンドデシルエーテル濃度 (mg/100ml)	反射光量
0.1	0.97
0.6	0.95
0.9	0.93
1.5	0.91
2.0	0.89

Example 3 To ion measurement implement tetrahydrofuran 10ml for zinc ion As a nonaqueous solubility giant molecule, 300mg (average degree of polymerization 1000) of polyvinyl chlorides, As an organic solvent, as 700mg of dioctyl sebacates, and a water soluble polymer Polyvinyl-pyrrolidone 10g, 2-(5-BUROMO-2-pyridyl azo)-5-diethylamino phenol 7.3mg (21micromol) was dissolved as an ionophore for zinc ion, and this was applied with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. When it is left in a room temperature and made to dry for 1 hour, the ion measurement implement which began to become muddy gradually and obtained the dark reddish-brown filmy material eventually and

which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the filmy material fixed on the transparence polypropylene sheet was obtained. Thus, if ion exchange water washes the obtained ion measurement implement, it can obtain only the organic phase part colored dark reddish-brown. As the example 1 showed, this organic phase part had taken three-dimension-network structure.

[0056] Example 4 The amount measuring device of reflected lights which shows the ion measurement implement produced in the measurement example 3 of zinc ion in drawing 2 was equipped, and 25micro of water solutions I of various zinc chloride concentration was dropped. The amount of reflected lights with a wavelength of 553nm was measured, and the absorbance was calculated on the basis of the amount of reflected lights at the time of zinc chloride concentration 0mM. The result was shown in drawing 3. Moreover, the time amount taken to become the fixed amount of reflected lights was also measured. Each required time amount was within the limits of 15 - 20 seconds.

[0057] 2-(5-BUROMO-2-pyridyl azo)-5-diethylamino phenol 7.3mg (21micromol) was dissolved in example of comparison 1 tetrahydrofuran 10ml as an ionophore 700mg of dioctyl sebacates, and for zinc ion as a nonaqueous solubility giant molecule as 300mg (average degree of polymerization 1000) of polyvinyl chlorides, and an organic solvent, and this was applied to it with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. The measurement implement which left it in the room temperature, was dried for 1 hour, and obtained the filmy material of transparence dark reddish-brown and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the filmy material fixed on the transparence polypropylene sheet was obtained.

[0058] By the same actuation as an example 4, various zinc chloride concentration water solutions were measured for this measurement implement. However, since the fixed amount of reflected lights was not reached even if 5 minutes passed after the water solution was dropped, it was not able to measure.

[0059] It dried, after ion exchange water washed the measurement implement produced in the example of comparison 2 example 3, and the water soluble polymer was removed, and the measurement implement which comes to dissolve a nonaqueous solubility macromolecule in the organic solvent of three-dimension-network structure was produced. By the same actuation as an example 4, various zinc chloride concentration water solutions were measured for this measurement implement. However, even after the water solution was dropped, the water solution did not permeate into the micropore of a measurement implement, and most change of a color was not seen. Since the fixed amount of reflected lights was not reached even if 5 minutes passed, it was not able to measure.

[0060] It was shown from examples 3 and 4, the example 1 of a comparison, and the example 2 of a comparison that it is necessary to fill up with the water soluble polymer for performing measurement in a short time at the organic interphase of three-dimension-network structure.

[0061] The tetrahydrofuran solution was applied on the transparence polypropylene sheet by the same presentation and the actuation as example of comparison 3 example 3, and it was made to dry quickly using a blower. The solution became cloudy rapidly. The measurement implement which was dried after that for 1 hour and obtained the filmy material of opaque dark reddish-brown and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the massive object fixed on the transparence polypropylene sheet was obtained.

[0062] By the same actuation as an example 4, various zinc chloride concentration water solutions were measured for this measurement implement. The result was shown in drawing 4. Since the response obtained since the part to which an about 0.1mm hole does not penetrate an aperture and light does not penetrate the film partly to a measurement implement by dropping of a water solution is made was very small, the unsuitable thing was shown in measurement.

[0063] Examples 3 and 4 and example 3 of a comparison, It was shown that it is the making [deposit / the deposit of the organic phase which becomes a difficulty water solubility difficulty volatility organic solvent list from a nonaqueous solubility macromolecule at least performs deposit actuation to the bottom of the condition which starts before a deposit of a water soluble polymer, and / fill up into the organic interphase of three-dimension-network structure with the

manufacture of the matter measurement implement of this invention by which the laminating was carried out on the base material]-water soluble polymer phase need.

[0064] Example 5 To ion measurement implement chloroform 10ml for sodium ion As a nonaqueous solubility giant molecule, as polystyrene 300mg and an organic solvent o-nitrophenyl octyl ether 600mg, As a water soluble polymer, polyethylene-glycol (average molecular weight 6000) 10g, They are 5, 11, 17, and 23-tetra--t-butyl as a sodium ionophore. - 25, 26, 27, 28 - Tetrakis (ethoxycarbonyl)-methoxy-calyx [4] allene 18.4mg (18.4micromol), 2 and 6-dibromo-3'-methoxy indophenol 7.1mg (18.4micromol) was dissolved as coloring matter, and this was applied with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. When it was made to leave and dry in a room temperature, the ion measurement implement which turbidity went up gradually and obtained the filmy material of a brown white color eventually and which cut this filmy material on 1cm square with the transparence polypropylene sheet, and the filmy material of a brown white color fixed on the transparence polypropylene sheet was obtained.

[0065] The amount measuring device of reflected lights which shows the ion measurement implement produced in the example 6 example 5 in drawing 2 was equipped, and 25micro (it prepares with a potassium hydroxide to pH5.5) of 2-morpholino ethane-sulfonic-acid 10mM buffer solutions I of various sodium chloride concentration was dropped. The amount of reflected lights with a wavelength of 670nm was measured, and the absorbance was calculated on the basis of the amount of reflected lights at the time of sodium chloride concentration 0mM. The result was shown in drawing 5. Drawing 5 shows that sodium ion can be measured between 10-0.1mM(s). Moreover, the time amount taken to become the fixed amount of reflected lights was also measured. Each required time amount was within the limits of 15 - 20 seconds.

[0066] To example of comparison 4 tetrahydrofuran 10ml, as a nonaqueous solubility giant molecule, 300mg (average degree of polymerization 1000) of polyvinyl chlorides, As an organic solvent, o-nitrophenyl octyl ether 600mg, They are 5, 11, 17, and 23-tetra--t-butyl as a sodium ionophore. - 25, 26, 27, 28 - Tetrakis (ethoxycarbonyl)-methoxy-calyx [4] allene 18.4mg (18.4micromol), 2 and 6-dibromo-3'-methoxy indophenol 7.1mg (18.4micromol) was dissolved as coloring matter, and this was applied with the doctor blade on the transparence polypropylene sheet at 600-micrometer thickness. It was made to leave and dry in a room temperature, and the measurement implement was obtained as a filmy material of uniform transparence brown.

[0067] Thus, the amount measuring device of reflected lights which shows the manufactured measurement implement in drawing 2 was equipped, and 25micro (it prepares with a potassium hydroxide to pH5.5) of 2-morpholino ethane-sulfonic-acid 10mM buffer solutions I of various sodium chloride concentration was dropped. Although the amount of reflected lights with a wavelength of 670nm was measured, even if 3 minutes passed, the fixed amount of reflected lights was not reached. It was not able to measure on the same conditions as an example 6 that a speed of response is slow.

[0068] It was shown that it can measure promptly using the ion measurement implement of this invention from an example 6 and the example 5 of a comparison.

[0069] The ion measurement implement for sodium ion was produced by the same actuation as an example 5 using the nonaqueous solubility macromolecule shown in seven to example 5-10 of comparison, and example 227 table 2, an organic solvent, a water soluble polymer, the same sodium ionophore as an example 5, and coloring matter. The amount measuring device of reflected lights which shows the produced measurement implement in drawing 2 was equipped, and 25micro (it prepares with a potassium hydroxide to pH5.5) of sodium chloride concentration 10 2-morpholino ethane-sulfonic-acid 10mM buffer solutions I of mM was dropped. The amount of reflected lights with a wavelength of 670nm was measured, and time amount until the amount of reflected lights at that time becomes fixed was measured. The result was shown in a table 2. The quick response was not obtained, even if missing in the nonaqueous solubility macromolecule, the organic solvent, or the water soluble polymer so that examples 5-10 might show. Moreover, it was shown that a quick response is obtained in the combination of various nonaqueous solubility macromolecules, an organic solvent, and a water soluble polymer from examples 7-22.

[0070]

[A table 2]

表 2

	非水溶性高分子	有機溶剤	水溶性高分子	応答時間 (秒)
比較例 5	なし	セバシン酸ジオクチル	ポリエチレングリコール	180 秒以上
比較例 6	〃	フタル酸ジオクチル	ポリエチレングリコール	180 秒以上
比較例 7	〃	〃	ポリビニルピロリドン	180 秒以上
比較例 8	〃	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	180 秒以上
比較例 9	ポリ塩化ビニル	なし	ポリエチレングリコール	反射量変化なし
比較例 10	〃	セバシン酸ジオクチル	なし	180 秒以上
実施例 7	〃	〃	ポリビニルピロリドン	17
実施例 8	〃	〃	ポリエチレングリコール	18
実施例 9	〃	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 10	〃	〃	ポリビニルピロリドン	20
実施例 11	〃	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	18
実施例 12	〃	〃	ポリビニルピロリドン	18
実施例 13	ポリメタクリルメチル	セバシン酸ジオクチル	ポリエチレングリコール	18
実施例 14	〃	〃	ポリビニルピロリドン	18

[0071]

[A table 3]

表 2 (続き)

	非水溶性高分子		水溶性高分子	応答時間 (秒)
実施例 15	ポリメタクリル酸メチル	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 16	〃	〃	ポリビニルピロリドン	20
実施例 17	〃	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	17
実施例 18	ポリスチレン	セバシン酸ジオクチル	ポリエチレングリコール	18
実施例 19	〃	〃	ポリビニルピロリドン	19
実施例 20	〃	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 21	〃	〃	ポリビニルピロリドン	19
実施例 22	〃	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	18

[Translation done.]

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2. *** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] This Fig. is the photograph which took a photograph of the structure of the organic phase after ion exchange water washes the matter measurement implement produced in the example 1 with the electron microscope.

[Drawing 2] It is drawing showing the configuration of the amount measuring device of reflected lights used in the example and the example of a comparison.

[Drawing 3] It is drawing showing the relation between the zinc ion concentration measured in the example 4, and the measured absorbance.

[Drawing 4] It is drawing showing the relation between the zinc ion concentration measured in the example 3 of a comparison, and the measured absorbance.

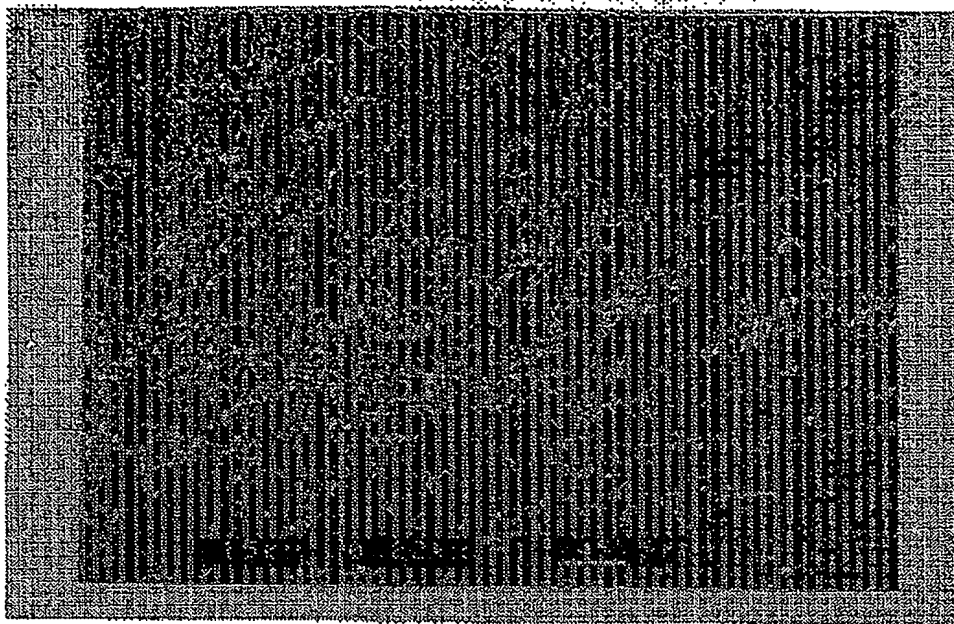
[Drawing 5] It is drawing showing the relation between the sodium ion concentration measured in the example 6, and the measured absorbance.

[Description of Notations]

- 1 Xenon Lamp
- 2 Condenser Lens
- 3 Optical Fiber for Floodlighting
- 4 Base (Quartz Plate)
- 5 White Reflecting Plate
- 6 Matter Measurement Implement of this Invention
- 7 Optical Fiber for Light-receiving
- 8 Flash Multi-Photometry Equipment (MCPD[by Otsuka Electronic Incorporated Company]-1000 Mold)
- 9 Data Processor (Otsuka Electronic Incorporated Company Make)

[Translation done.]

写真用代用図面



写真

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(71) 出願人 000003182

株式会社トクヤマ

山口県徳山市御影町1番1号

(72) 発明者 渡辺 伸

山口県徳山市御影町1番1号 株式会社ト

クヤマ内

(72) 発明者 榎 徹

山口県徳山市御影町1番1号 株式会社ト

クヤマ内

(54) 【発明の名称】 物質測定具、その製造方法及びイオン測定具

(57) 【要約】

【構成】 セバシン酸ジオクチル、*o*-ニトロフェニルオクチルエーテル等の難水溶性難揮発性有機溶剤にポリ塩化ビニル、ポリスチレン等の非水溶性高分子を溶解してなる有機相とポリエチレングリコール、ポリビニルピロリドン等の水溶性高分子相からなり、水溶性高分子相が3次元網目状構造の有機相間に充填されてなる物質測定具。

【効果】 本発明の物質測定具は、水溶性高分子相が、難水溶性難揮発性有機溶剤に非水溶性高分子を溶解してなる3次元網目状構造の有機相間に充填されてなるために、測定試料は速やかに物質測定具全体に浸透することができ、また有機相の厚みが小さいために測定試料から有機相への測定対象物質の移動が速やかに行われるという特長を有する。そのため、従来煩雑な操作が必要であった抽出分離操作を簡便な操作で、かつ短時間に行うことができ、その結果被測定対象物質を迅速、簡便に測定できるという特長を有する。

【特許請求の範囲】

【請求項1】 (A) 難水溶性難揮発性有機溶剤に非水溶性高分子を溶解してなる有機相と(B)水溶性高分子相からなり、(A)有機相が3次元的網目状構造をなし該(A)有機相間に(B)水溶性高分子相が充填されていることを特徴とする物質測定具。

【請求項2】 請求項1記載の物質測定具が支持体上に積層されてなる物質測定具。

【請求項3】 難水溶性難揮発性有機溶剤、非水溶性高分子および水溶性高分子を有機溶媒に溶解してなる溶液を支持体上に塗布し、次いで難水溶性難揮発性有機溶剤および非水溶性高分子からなる有機相の析出が水溶性高分子の析出よりも先に始まるか或は同時に析出する条件下に有機溶媒を蒸発させることを特徴とする請求項2記載の物質測定具の製造方法。

【請求項4】 (A) 難水溶性難揮発性有機溶剤に、イオンと錯体形成して検出可能な応答を示すイオノフォアまたはイオンと錯体形成するイオノフォアと該錯体形成に相互反応して検出可能な応答を示す色素、並びに非水溶性高分子を溶解してなる有機相と(B)水溶性高分子相からなり、(A)有機相が3次元的網目状構造をなし該(A)有機相間に(B)水溶性高分子相が充填されていることを特徴とするイオン測定具。

【請求項5】 請求項4記載のイオン測定具が支持体上に積層されてなるイオン測定具。

【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は溶液中の様々な物質の検出、定量に使用することができる物質測定具並びにこれを利用したイオン測定具に関する。

【0002】

【従来の技術】分析化学の分野において、溶液中、特に水溶液中の様々な物質を測定し医療、生産活動、環境保全などに役立てることが行われている。たとえば人体の血清中、尿中のイオン濃度を測定することにより患者の病態に関しての有益な情報が得られることが知られている。また、企業の生産活動の結果として排出される廃液中の有害な重金属類の監視、生活排水などに含まれる界面活性剤の濃度の測定は環境保全に必要とされている。その他例を挙げれば枚挙がないが、溶液中の物質の測定は我々の生活には欠かせないこととなっている。

【0003】溶液中の物質の測定方法は、その測定対象物質により異なり、数多くの方法が存在する。その中に、測定対象物質によって誘起される吸収あるいは蛍光スペクトルの変化を測定する、比色法と呼ばれる方法がある。比色法においては水溶液中の測定対象物質を有機溶媒中に抽出分離し、有機溶媒に移動した測定対象物質と、あらかじめ水中あるいは有機溶媒中に添加しておいた発色試薬との相互作用により生じる吸収あるいは蛍光スペクトルの変化量から定量を行うことが多い。

【0004】抽出分離とは、2種の液体の一方からもう一方へとある物質を分離させるために行う操作である。一般的には互いに交じり合わない2種の溶液を分液ロートに入れ、これを振り混ぜ、その後測定対象物質が移動した側の溶液のみを取り出すという操作を行う。そのため抽出分離操作が必要となる比色法の測定では、抽出分離のための専用の用具が必要となる。この欠点を補うため、有機溶媒のかわりに非水溶性高分子を添加した難水溶性難揮発性有機溶剤からなる膜を使用することが知られている。たとえば、「分析化学」第40巻(1991年)227~231ページにおいて、一価の銅イオンと特異的に錯体を形成し発色するバソクブロインを非水溶性高分子と難水溶性難揮発性有機溶剤に溶解させた膜を使用し、この膜を未知濃度の一価の銅イオンを含有する水溶液に接触させることにより銅イオンを膜中に抽出分離して膜中のバソクブロインを発色せしめて、膜の吸光度から銅イオン濃度を測定する方法が述べられている。しかし、抽出分離操作においては水溶液中の測定対象物質が難水溶性難揮発性有機溶剤中に移動するという物質移動が必須であるために、測定対象物質の難水溶性難揮発性有機溶剤中での拡散が律速になり、測定にかかる時間が非常に長いという短所があった。

【0005】測定対象物質の移動により吸収あるいは蛍光スペクトルの変化を生じる原理の比色法によるイオンの分析法が特開昭59-211864号公報、特開昭59-231452号公報、特開昭60-194360号公報に述べられている。これらは種々のイオノフォアおよび、レポータ物質と呼ばれる色素を含む難水溶性難揮発性有機溶剤からなるイオン濃度測定具である。これらの測定具では測定対象物質であるイオンを含む測定試料水溶液と接触させることにより、イオンの膜内への移動が起き、色素の吸光スペクトルあるいは蛍光スペクトルに変化を生じる。この方法においてもイオンが膜内で拡散する速度が律速となることが知られている。膜厚を数 μm 程度に薄くすることが有効な手法であることが知られている。しかし、発色変化の速度は大きくなる半面、得られる発色変化量が小さくなるという欠点を有するため実用的ではない。

【0006】上記の欠点を克服するために様々な手法が開発されてきた。たとえば特開昭60-194360号公報に種々のイオノフォアおよび、レポータ物質と呼ばれるpH感受性色素を含む難水溶性難揮発性有機溶剤を多孔質担体に担持させたイオン濃度測定具が記載されている。ろ紙のような多孔質担体の表面にイオノフォアおよび、レポータ物質を含む難水溶性難揮発性有機溶剤を薄く被覆させることにより、発色変化の速度と発色変化量の問題を共に解決しようという試みであった。この手法によりある程度の解決をすることはできた。しかしこれら多孔質担体を利用する測定具は、製造を行う際に多孔質担体を支持体上に正確な位置で固定するという工程を必要とし、大量かつ均一に製造を行うのが困難、あるいは非常

にコストがかかるという欠点をなお有していた。そのため多孔質担体を使用せずに同様の効果を持つ手法の開発が望まれていた。

【0007】

【発明が解決しようとする課題】実用上十分な応答速度と応答量を兼ね備えた測定を行うことのできる物質測定具、例えば比色法に使用したときには実用上十分な発色速度と発色変化量を兼ね備えた測定を行うことのできる物質測定具、並びにこれを利用したイオン測定具の開発を目的とする。

【0008】

【課題が解決するための手段】本発明者らは、かかる課題を解決しうる物質測定具を開発すべく鋭意研究を重ねた。その結果、難水溶性難揮発性有機溶剤に非水溶性高分子を溶解してなる有機相と水溶性高分子相が特定の構造を取ったときに上記課題を解決するに十分な性能を発揮することを見いだした。

【0009】すなわち本発明は、(A) 難水溶性難揮発性有機溶剤に非水溶性高分子を溶解してなる有機相と(B) 水溶性高分子相からなり、(A)有機相が3次元的網目状構造をなし該(A)有機相間に(B)水溶性高分子相が充填されていることを特徴とする物質測定具であり、更に該物質測定具が支持体上に積層されてなる物質測定具である。

【0010】他の発明は、(A) 難水溶性難揮発性有機溶剤に、イオンと錯体形成して検出可能な応答を示すイオンフォアまたはイオンと錯体形成するイオンフォアと該錯体形成に相互反応して検出可能な応答を示す色素、並びに非水溶性高分子を溶解してなる有機相と(B)水溶性高分子相からなり、(A)有機相が3次元的網目状構造をなし該(A)有機相間に(B)水溶性高分子相が充填されていることを特徴とするイオン測定具であり、更に該イオン測定具が支持体上に積層されてなるイオン測定具である。

【0011】更に他の発明は、 難水溶性難揮発性有機溶剤、非水溶性高分子および水溶性高分子を有機溶媒に溶解してなる溶液を支持体上に塗布し、次いで難水溶性難揮発性有機溶剤および非水溶性高分子からなる有機相の析出が水溶性高分子の析出よりも先に始まるか或は同時に析出する条件下に有機溶媒を蒸発させることを特徴とする上記物質測定具の製造方法である。

【0012】本発明の物質測定具を構成する有機相は基本的に難水溶性難揮発性有機溶剤と非水溶性高分子から成る。

【0013】上記難水溶性難揮発性有機溶剤（以下、単に有機溶剤という）とは、水に難溶かつ高沸点であり、イオン化し得る官能基を有しない、常温で液状の有機化合物のことを指す。該有機溶剤は測定試料中の測定対象物質を抽出し、また測定に必要な色素等を溶解しておくために使用される。具体的には沸点が摂氏100度以上で

あり、水11に対して溶解することのできる有機溶剤が1g以下程度であることが安定した測定をするために望ましい。

【0014】好適に使用される有機溶剤を具体的に挙げれば、アジピン酸ジブチル、フタル酸ジオクチルなどの高沸点エステル類、*o*-ニトロフェニルオクチルエーテル、ジフェニルエーテルなどの高沸点エーテル類がある。

【0015】非水溶性高分子としては上記有機溶剤に溶解することができ、かつ水に不溶の高分子を用いる。該非水溶性高分子は上記有機溶剤に溶解して、物質測定具としての使用に適した粘度、物理的強度を付与する。

【0016】好適に使用される非水溶性高分子の例を挙げればポリ塩化ビニル、エチレン-塩化ビニル共重合体などのポリ塩化ビニル系ポリマー、塩化ビニル-塩化ビニリデン共重合体、塩化ビニリデンなどの塩化ビニリデン系ポリマー、ポリメタクリル酸メチルなどのメタクリル酸エステル系ポリマー、ポリスチレンなどのスチレン系ポリマー等がある。非水溶性高分子の重合度については特に限定されることがなく使用することができるが、一般に平均重合度500以上のものを用いると有機溶剤が自己支持性を示すようになり好ましい非水溶性高分子の有機溶剤中の濃度は、その溶解度の上限以下で、かつ有機溶剤が自己支持性を示す範囲であると物質測定具としたときの取り扱いが容易になり好適である。その濃度範囲は使用する非水溶性高分子、有機溶剤の種類により異なるが、一般に有機溶剤に対して非水溶性高分子を5～100重量%溶解させて用いるのが好適である。非水溶性高分子の有機溶剤への溶解方法は特に限定されず、通常有機溶剤中へ非水溶性高分子を徐々に添加しながら溶解させる。

【0017】本発明を構成する水溶性高分子相は基本的に水溶性高分子から成る。該水溶性高分子は3次元的網目状構造の有機相の間を測定試料が迅速に浸透するのを促進するために使用される。

【0018】水溶性高分子とは水に溶解することができる高分子のことであり、水溶性高分子はその溶解度が50wt%以上のものを用いることが望ましい。水溶性高分子が存在しないと測定試料は有機溶剤の隅々にまでいきわたることができないため、物質測定具を測定に用いた時に要する測定時間が長くなる。水溶性高分子には有機溶剤に難溶のものを用いても本発明の効果を得ることができるが、その場合には測定試料と本発明の物質測定具を接触させた際に著しい変形を伴い、安定した測定ができない場合がある。また、物質測定具として使用する際の取り扱いの容易さを勘案すると、室温中で固体状態となる重合度のものを用いることが望ましい。水溶性高分子の量は有機溶剤、非水溶性高分子を含めた全体量に対し500～100重量%の範囲であれば、適切な物理的強度を

保ちつつ測定試料の浸透を促進するのに望ましい。

【0019】好適に使用できる水溶性高分子の例を挙げれば、ポリエチレングリコール、ポリプロピレングリコールなどのポリエーテル系高分子化合物、ポリビニルピロリドン、ポリビニルアルコール、ポリアクリルアミドなどがある。

【0020】本発明の有機相が3次元網目状構造であることは水溶性高分子を水で洗い流した後に残る部分を、電子顕微鏡等で観察することにより確認することができる。

【0021】水溶性高分子相を、3次元網目状構造の有機相間に充填状態で存在させる方法には様々の方法を使用することができるが、最も好適に使用される方法は次のとおりである。

【0022】即ち、有機溶剤、非水溶性高分子および水溶性高分子をすべて有機溶媒に溶解させ、その後に支持体上に塗布し乾燥させる。有機溶媒の蒸発に伴い、有機溶剤に非水溶性高分子を溶解してなる有機相と水溶性高分子相のミクロ相分離が起き、水溶性高分子相が3次元網目状構造の有機相に充填された状態で析出する。

【0023】上記方法に使用する有機溶媒としては有機溶剤、非水溶性高分子、水溶性高分子をすべて溶解するものであればなんら制限なく用いることができる。選択した有機溶剤、非水溶性高分子、水溶性高分子の組み合わせにより使用できる有機溶媒は異なるが、その例を挙げればテトラヒドロフランなどのエーテル類、ジクロロメタン、クロロホルム、ジクロロエタンなどのハロゲン化アルキル類がある。有機溶媒の使用量は選択した有機溶剤、非水溶性高分子、水溶性高分子の溶解度により異なるため一概に規定することはできないが、析出を促すため有機溶剤、非水溶性高分子、水溶性高分子のいずれかを有機溶媒と同量以上とするのが一般的である。

【0024】上記の析出法では有機溶剤および非水溶性高分子からなる有機相の析出が水溶性高分子相の析出よりも先に始まるか或は同時に析出する場合に均一で良好な構造を得ることができる。有機相の析出が先に始まる条件で製造した場合がより好適である。逆に、水溶性高分子の析出が有機相の析出よりも先に始まる場合には、得られる物質測定具は不均一かつ空隙の多いものとなり、比色法による測定には適さない。

【0025】有機相と水溶性高分子相の析出順序の制御は、両相を溶解させる有機溶媒の有機相と水溶性高分子相に対する溶解性を制御することにより行うことが可能である。この溶解性の制御は一般に有機溶媒の極性により制御される。一般に有機相を構成する有機溶剤および非水溶性高分子は極性の低い有機溶媒に溶解し易く、逆に水溶性高分子は極性の高い有機溶媒に溶解し易いという性質を有する。そのため、極性の高い有機溶媒を使用すれば、溶解度の低い有機相の析出が先に始まる。よって、有機溶剤および非水溶性高分子からなる有機相の析

出を水溶性高分子相の析出よりも先に始めさせることができる。

【0026】有機溶媒の極性は2種以上の有機溶媒あるいは水を混合することにより制御することができる。好適に使用される例を示せば、約0.1%の水を添加したテトラヒドロフラン、1%メタノールを添加したクロロホルム等がある。また、有機相にイオノフォア、色素を溶解させた場合も、何等問題なく同様の方法で析出の順序を制御することができる。

10 【0027】良好な構造の物質測定具を得る具体的な例を挙げれば、有機溶剤としてo-ニトロフェニルオクチルエーテル、非水溶性高分子としてポリメタクリル酸メチル、水溶性高分子としてポリエチレングリコール（平均分子量約6000）を用い、これらをテトラヒドロフランに溶解し、ガラス板上に塗布し、室温中で放置して乾燥させることにより作製することができる。このときにテトラヒドロフラン中に微量の水分が存在すると、ポリエチレングリコールの析出が遅くなり、再現性良く上記の構造を得ることができる。有機溶剤および非水溶性高分子からなる有機相の析出が水溶性高分子相の析出よりも先に始まることは、着色した有機相あるいは色素を溶解させた有機相を用いることにより、着色した析出物が析出することによって知ることができる。

20 【0028】有機溶剤に、イオンと錯体形成して検出可能な応答を示すイオノフォアを溶解してなる有機相をイオン濃度の測定に用いる原理および方法は公知であり「分析化学」第40巻（1991年）227～231ページにその記述がある。また、有機溶剤に、イオンと錯体形成するイオノフォアと該錯体形成に相互反応して検出可能な応答を示す色素、並びに非水溶性高分子を溶解してなる有機相をイオン濃度の測定に用いる原理、方法も公知であり特開昭59-211864号公報、「アナリティカルサイエンス」(ANALYTICAL SCIENCE)1990年715～720ページなどにその記述がある。本発明にて用いられるイオノフォアおよび色素はこれらの文献にて述べられているものを何ら問題なく使用することができる。

30 【0029】本発明において、イオノフォアとは特定のカチオンと錯体形成をする性質をもった化合物である。特定の金属イオンと錯体を形成する能力を持ち、該金属イオンと錯体を形成することにより電子軌道状態に変化を生じ、錯体の吸収スペクトルあるいは蛍光スペクトルに変化を生じる色素は、本発明のイオンと錯体形成して検出可能な応答を示すイオノフォアとして使用することができる。このようなイオノフォアとしてはニトロソアミノフェノール類、ジエチルアミノフェノール類、ボルフィリン類、フェノキサジン類などがある。

40 【0030】また、本発明のイオンと錯体形成するイオノフォアと当該錯体形成に相互反応して検出可能な応答を示す色素の組合せにて使用することのできるイオノフォアとしては、公知の数種の環状ペプチド化合物、環状

または非環状ポリエーテル誘導体、ポタンド誘導体、カリックスアレン誘導体などがある。

【0031】イオンと錯体を形成して検出可能な応答を示すイオノフォアの好適に用いることのできる具体例を挙げれば、カリウムイオンに対しては3'-ニトロ-4'-

(2,4,6-トリニトロフェニルアミノ)ベンゾ-18-クラウン-6、4'- (2,6-ジニトロ-4-トリフルオロメチルフェニル)アミノベンゾ-15-クラウン-5等が、銅イオンに対してはバクブロン等が、鉄イオンに対しては2-ニトロソ-5-N,N-ジプロピルアミノフェノール等のニトロソアミノフェノール類が、ニッケル、亜鉛イオンに対しては2-(5-ブromo-2-ピリジルアゾ)-5-ジエチルアミノフェノール等のジエチルアミノフェノール類が、水素イオンに対しては9-(ジエチルアミノ)-5-オクタデカノイルイミノ-SH-ベンゾ[a]フェノキサジンなどのフェノキサジン類、[5-オクタデカノイルキシ-2-(4-ニトロフェニルアゾ)フェノール]などのアゾフェノール類がある。

【0032】一方、イオンと錯体形成するイオノフォアと当該錯体形成に相互反応して検出可能な応答を示す色素の組合せにて使用されるイオノフォアの好適に用いることができる具体例をあげれば、リチウムイオノフォアとしては6,6-ジベンジル-1,4,8,11-テトラオキサシクロテトラデカン、2,9-ジブチル-1,10-フェナントロリン等が；ナトリウムイオノフォアとしては5,11,17,23-テトラ-ヒプチル-25,26,27,28-テトラキス(エトキシカルボニル)-メトキシ-カリックス[4]アレン、ビス[(12-クラウン-4)メチル]メチルドデシルマロネート等が；カリウムイオノフォアとしてはバリノマイシン、2,3-ナフト-1,4,7,10,13-ペンタオキサシクロペンタデカ-2-エン、ビス[(ベンゾ-15-クラウン-5)-4'-メチル]ビメレート、バリノマイシン等が；カルシウムイオノフォアとしてはジエチル-N,N'-[(4R,5R)-4,5-ジメチル-1,8-ジオキソ-3,6-ジオキサオクタメチレン]ビス(12-メチルアミノドデカノエート)等が；マグネシウムイオノフォアとしてはN,N'-ジヘプチル-N,N'-ジメチルアスパルトアミド等が、銅イオンに対してベンゾ-15-チアクラウン-4-エーテル等がある。

【0033】もちろん、この他にもカチオンと錯体を形成をすることが知られている化合物は存在し、それらも使用することができる。イオノフォアに関する文献としてはYoshihisa Inoue, George W. Gokel編「Cation Binding by Macrocycles」(MARCEL DEKKER INCORPORATION 1990年)、武田裕行編著「機能性大環状化合物の分析化学への応用」(株式会社アイピーシー1990年発行)アナリティカルケミストリー (ANALYTICAL CHEMISTRY) 60巻2013-2016ページ(1988年)、「分析化学」23巻1412-1430ページ(1974年)などが挙げられる。

【0034】上述のイオンと錯体形成するイオノフォアと該錯体形成に相互反応して検出可能な応答を示す色素

(以後リポータ色素と記す)としてはpH指示薬、あるいはpH感受性色素と呼ばれる一連の色素がある。

【0035】本発明において使用されるリポータ色素は有機溶剤に可溶化されて含有されるため、水溶性が著しく小さなものが使用される。その入手の容易さ、イオン測定具として使用したときの発色応答の程度から、pKaが5~10のリポータ色素が好適に使用される。

【0036】リポータ色素を具体的に示せば、フェノールフタレイン、テトラブromoフェノールフタレイン等のフェノールフタレイン誘導体、2,6-ジクロロインドフェノール、2,6-ジブromo-3'-メチルインドフェノール等のインドフェノール誘導体、ピクトリアビュアブルー等のトリフェニルメタン色素誘導体等がある。

【0037】イオノフォアおよびリポータ色素はその溶解度の上限まで有機溶剤に溶解させることができる。このとき、イオノフォアとリポータ色素のモル比はその原理より1:1であることが好適であることが知られている。一般的にはイオノフォアとリポータ色素の有機溶剤中での濃度は、測定を行ったときの感度を勘案してそれぞれ0.1~200mMの範囲であることが望ましい。

【0038】本発明において、有機溶剤に非水溶性高分子を溶解してなる有機相が3次元的網目状構造であるとは、有機相がスポンジ、海綿のように3次元的に網目状構造を有していることを指す。同様の構造を持つ物質は吸水性物質あるいは濾過分離用として、異なる材質で様々なものが用いられている。3次元的網目状の構造を持つ非水溶性高分子からなる物質の製法は特開平2-302449、特開昭58-104940、特開昭47-4593などに記載されている。これらの従来の3次元的網目状構造の物質は高い強度を実現するために非水溶性高分子を主体とし、一般には有機溶剤を添加することはない。また、有機溶剤が添加されていても水溶性高分子を充填したものは知られていない。本発明の3次元的網目状構造の有機溶剤に非水溶性高分子を溶解してなる有機相は、抽出操作を行うための有機溶剤を主体とするものであり、非水溶性高分子は適度な粘度と物理的強度を付与するために添加される、という根本的な違いがある。

【0039】本発明において、3次元的網目状構造の有機相間に水溶性高分子相が充填されていることにより、測定試料は速やかに3次元的網目状構造の有機相の微細孔内に浸透する。また、有機相は3次元的網目状構造であるため数μm以下の厚みすることができ、その結果、速やかに測定対象物質の有機相内への移動を完了しうる。また3次元的網目状構造の有機相の、水溶性高分子を取り除いた時にできる微細孔の孔径は500μm以下である場合に均一な構造を取りうる。均一な構造をとることによりより正確な測定を可能とし、好ましい。

【0040】本発明の物質測定具はそのままで測定に用いることができるが、測定の際の操作性、pH緩衝剤等試薬との組み合わせでの使用などを勘案して支持体上に

積層することができる。積層の方法としては特に限定することなく公知の方法を用いることができる。好適に用いられる例を挙げると、本発明の物質測定具をあらかじめ作製しておき適切な接着剤、両面テープなどで接着する方法。有機溶剤、非水溶性高分子および水溶性高分子を有機溶媒に溶解してなる溶液を支持体上に塗布し、前述の通り有機溶剤に非水溶性高分子を溶解してなる有機相が3次元的網目状構造に析出する条件下で乾燥させる方法などがある。

このようにして作製した物質測定具を用い、種々の物質の濃度の測定を行うことができる。以下に具体例をもって説明をする。

【0041】現在、水溶液中の界面活性剤の界面活性剤の濃度の測定は、界面活性剤と特異的に結合あるいは錯体形成する着色物質を水溶液中に添加し、界面活性剤と着色物質とともに水溶液中から非水溶性の抽出液へと抽出し、その後抽出液の着色度を測定することにより行われている。このような方法は日本工業規格に示されており（JIS K 0102 -1986 工場排水試験方法）、メチレンブルー吸光度法、テトラチオシアノコバルト（II）酸吸光度法などが記載されている。これらの方法はいずれも2種類の液を容器にいれてから、振り混ぜて抽出するという操作を伴うため、大変な時間と労力を要するという欠点を持っている。

【0042】本発明の物質測定具を用いて、同様の原理で上記測定を行うことができる。メチレンブルー吸光度法を例にとれば以下のとおりである。すなわち、一定量の測定試料水溶液に一定量のメチレンブルーを添加し、これを本発明の物質測定具に滴下すると、物質測定具の有機相中にメチレンブルーと陰イオン界面活性剤のイオン対が抽出分離される。その後イオン交換水等を用いて物質測定具に付着した測定試料水溶液を洗い流し、残った有機相の着色度を測定する。着色度の測定は目視あるいは透過光量、反射光量の測定により行うことができる。このように、本発明の物質測定具を用いると簡便な操作で特定物質の測定を行うことができる。

【0043】また、本発明の、有機溶剤にイオンと錯体形成して検出可能な応答を示すイオノフォア並びに非水溶性高分子を溶解してなる3次元的網目状構造の有機相間に水溶性高分子相が充填されてなるイオン測定具を用いる場合には、以下のようにして測定を行うことができる。

【0044】銅イオン用のイオノフォアとしてバソクブロインを使用する場合を例にとれば次のとおりである。有機溶剤にバソクブロインおよび非水溶性高分子を溶解させてなる3次元的網目状構造の有機相間に水溶性高分子相が充填されてなるイオン測定具に銅イオンを含有した測定試料水溶液を滴下する。測定試料水溶液は速やかにイオン測定具全体に浸透し、銅イオンはイオン測定具の有機相内に存在するバソクブロインにより有機相内へ

と抽出される。有機相は3次元的網目状構造であるため非常に膜厚が薄くなっており、この抽出は速やかに行われる。バソクブロインは抽出された銅イオンとの錯体形成によりその吸収スペクトルに変化を生じる。一定時間経過後、イオン交換水等を用いてイオン測定具に付着した測定試料水溶液を洗い流し、残った有機相の着色度を測定することにより、測定試料水溶液に含有されている銅イオンの濃度を知ることができる。従来の膜による抽出法では完全に応答が終了するのに数時間かかったが、本発明のイオン測定具では数分以内で応答を終了させることができる。

【0045】さらに、本発明の有機溶剤にイオンと錯体形成するイオノフォア、リポータ色素および非水溶性高分子を溶解してなる3次元的網目状構造の有機相間に、水溶性高分子相が充填されてなるイオン測定具とした場合には、以下のようにして測定を行うことができる。

【0046】カリウム用のイオノフォアとしてバリノマイシン、リポータ色素として2,6-ジブromo-3'-メトキシインドフェノールを用い、水溶液中のカリウムイオンを測定する場合を例にとれば、以下のとおりである。有機溶剤にバリノマイシン、2,6-ジブromo-3'-メトキシインドフェノールおよび非水溶性高分子を溶解してなる3次元的網目状構造の有機相間に、水溶性高分子相が充填されてなるイオン測定具に測定試料水溶液を滴下する。このとき、測定試料水溶液のpHはpH緩衝剤などを用いて一定に保っておく必要がある。測定試料水溶液は速やかにイオン測定具全体に浸透し、カリウムイオンはイオン測定具の有機相内に存在するバリノマイシンにより有機相内へと抽出分離され、それと同時に2,6-ジブromo-3'-メトキシインドフェノールが水素イオンを測定試料水溶液中に放出して発色する。測定試料水溶液中のカリウムイオンが多いほど膜中に抽出されるカリウムイオンは増加し、それに伴い発色するリポータ色素の量も増加する。結果として、測定試料水溶液中のカリウムイオンの量に応じた測定具の着色が観察され、イオン測定具の着色度からカリウムイオンの濃度を知ることができる。有機相は3次元的網目状構造であるため非常に膜厚が薄くなっており、この抽出および放出は速やかに行われ、測定に要する時間は短いという特長を有する。

【0047】もちろん、本発明の物質測定具は、有機溶剤または非水溶性の有機溶媒を用いて、抽出あるいはイオン交換を行う他の公知の測定原理にも応用することができる。いずれの場合も短時間かつ簡便な操作で測定を行うことができるという特長を有している。

【0048】

【発明の効果】本発明の物質測定具は、水溶性高分子相が、難水溶性難揮発性有機溶剤に非水溶性高分子を溶解してなる3次元的網目状構造の有機相間に充填された基本構造を持つために、測定試料は速やかに物質測定具全体に浸透することができ、また有機相の厚みが小さいた

めに測定試料から有機相への測定対象物質の移動が速やかに行われるという特長を有する。そのため、従来煩雑な操作が必要であった抽出分離操作を簡便な操作で、かつ短時間に行うことができるという特長を有する。

【0049】従って、緊急性の高い測定の場合、すなわち救急医療の現場や手術室における血中イオン濃度測定においては多大なる利益をもたらす。また、一般の臨床検査においても限られた時間内の血清あるいは尿検体の測定数を増大せしめることとなり、作業の効率化および診断の円滑化をもたらす。また、煩雑な操作を有するために従来は実験室内のみ行われていたような測定も、測定試料採取現場で行うことができ、測定結果を迅速に知ることができるようになる。

【0050】さらに本発明の物質測定具は、従来の多孔質担体を使用しないため、簡単な工程にて製造することができる。そのため安価に製造することができるという特長を有する。以上の点で、本発明の工業的価値は極めて大きい。

【0051】

【実施例】本発明をさらに具体的に説明するため、以下に実施例および比較例を挙げて説明するが、本発明はこれらの実施例に限定されるものではない。

【0052】実施例1

テトラヒドロフラン10mlに、非水溶性高分子としてポリ塩化ビニル（平均重合度1000）300mg、有機溶剤としてセバシン酸ジオクチル700mg、水溶性高分子としてポリ

表 1

ヘプタオキシエチレンドデシルエーテル濃度 (mg/100ml)	反射光量
0.1	0.97
0.6	0.95
0.9	0.93
1.5	0.91
2.0	0.89

実施例3 亜鉛イオン用のイオン測定具

テトラヒドロフラン10mlに、非水溶性高分子としてポリ塩化ビニル（平均重合度1000）300mg、有機溶剤としてセバシン酸ジオクチル700mg、水溶性高分子としてポリビニルピロリドン10g、亜鉛イオン用のイオンフォアとして2-(5-ブromo-2-ピリジルアゾ)-5-ジエチルアミノフェノール7.3mg (21μmol)を溶解し、これを透明ポリプロピレンシート上にドクターブレードにて600μm厚に塗布した。室温中で放置し1時間乾燥させると、徐々に濁り始め最終的に赤褐色の膜状物を得た、この膜状物を透明ポリプロピレンシートと共に1cm角に切断し、膜状物が透明ポリプロピレンシートに固着したイオン測

* エチレングリコール（平均分子量4000）10gを溶解し、これを透明ポリプロピレンシート上にドクターブレードにて600μm厚に塗布した。室温中で放置し1時間乾燥させると、徐々に白濁し始め最終的に白色の膜状物を得た、この膜状物を透明ポリプロピレンシートと共に1cm角に切断し、白色の膜状物が透明ポリプロピレンシートに固着した物質測定具を得た。

【0053】この物質測定具をイオン交換水100ml中に3分間浸せし、透明ポリプロピレンシート上に残った有機相を電子顕微鏡にて観察した。この際に撮影された写真を第1図に示す。3次元の網目状構造物が有機相により形成されていることが確認された。

【0054】実施例2 非イオン性界面活性剤の分析
非イオン性界面活性剤であるヘプタオキシエチレンドデシルエーテルの各種濃度の水溶液10mlに0.30Mテトラチオシアノコバルト(II)酸アンモニウム水溶液1.5mlを加え、1分間攪拌した。この水溶液を実施例1の物質測定具に20μl滴下し、1分後に物質測定具をイオン交換水約2mlで洗浄した。物質測定具の330nmでの反射光量を第2図に示す反射光量測定装置にて測定した結果を表1に示した。本発明の物質測定具を用いて簡便な操作にて非イオン性界面活性剤の定量を行うことができることが示された。

【0055】

【表1】

定具を得た。このようにして得られたイオン測定具はイオン交換水にて洗浄すると赤褐色に着色した有機相部分のみを得ることができる。実施例1にて示したようにこの有機相部分は3次元の網目状構造をとっていた。

【0056】実施例4 亜鉛イオンの測定

実施例3にて作製したイオン測定具を第2図に示す反射光量測定装置に装着し、各種塩化亜鉛濃度の水溶液25μlを滴下した。波長553nmでの反射光量を測定し、塩化亜鉛濃度0mMの時の反射光量を基準として吸光度を計算した。結果を第3図に示した。また一定の反射光量になるまでに要した時間をも測定した。要した時間はいずれも15～20秒の範囲内であった。

【0057】比較例1

テトラヒドロフラン10mlに、非水溶性高分子としてポリ塩化ビニル（平均重合度1000）300mg、有機溶剤としてセバシン酸ジオクチル700mg、亜鉛イオン用のイオノフォアとして2-（5-プロモ-2-ピリジルアゾ）-5-ジエチルアミノフェノール7.3mg（21 μ mol）を溶解し、これを透明ポリプロピレンシート上にドクターブレードにて600 μ m厚に塗布した。室温中で放置し1時間乾燥させ、透明赤褐色の膜状物を得た、この膜状物を透明ポリプロピレンシートと共に1cm角に切断し、膜状物が透明ポリプロピレンシートに固着した測定具を得た。

【0058】この測定具を実施例4と同様の操作で、各種塩化亜鉛濃度水溶液の測定を行った。しかし、水溶液を滴下した後、5分経過しても一定の反射光量に達しなかったため、測定を行うことができなかった。

【0059】比較例2

実施例3で作製した測定具をイオン交換水で洗浄した後乾燥して水溶性高分子を取り除き、3次元的網目状構造の有機溶剤に非水溶性高分子を溶解してなる測定具を作製した。この測定具を実施例4と同様の操作で、各種塩化亜鉛濃度水溶液の測定を行った。しかし、水溶液を滴下した後も水溶液は測定具の微細孔中には浸透せず、色の変化はほとんど見られなかった。5分経過しても一定の反射光量に達しなかったため、測定を行うことができなかった。

【0060】実施例3、4、比較例1、比較例2より、短時間での測定を行うには水溶性高分子が3次元的網目状構造の有機相間に充填されている必要があることが示された。

【0061】比較例3

実施例3と同じ組成および操作で透明ポリプロピレンシート上にテトラヒドロフラン溶液を塗布し、送風機を用いて急速に乾燥させた。溶液は急激に白濁した。その後1時間乾燥させ、不透明赤褐色の膜状物を得た、この膜状物を透明ポリプロピレンシートと共に1cm角に切断し、塊状物が透明ポリプロピレンシートに固着した測定具を得た。

【0062】この測定具を実施例4と同様の操作で、各種塩化亜鉛濃度水溶液の測定を行った。結果を第4図に示した。水溶液の滴下により測定具に0.1mm程度の穴がいくつか開き、光が膜を透過しない部分ができてしまうために、得られる応答は非常に小さいため、測定には不適であることが示された。

【0063】実施例3、4および比較例3より、支持体上に積層された本発明の物質測定具の製造では、少なくとも難水溶性難揮発性有機溶剤並びに非水溶性高分子からなる有機相の析出が水溶性高分子の析出よりも先に始まる条件下に析出操作を行って、3次元的網目状構造の有機相間に水溶性高分子相を充填させることが必要であることが示された。

【0064】実施例5 ナトリウムイオン用のイオン測定具

クロロホルム10mlに、非水溶性高分子としてポリスチレン300mg、有機溶剤としてo-ニトロフェニルオクチルエーテル600mg、水溶性高分子としてポリエチレングリコール（平均分子量6000）10g、ナトリウムイオノフォアとして5,11,17,23-テトラ- ϵ -ブチル-25,26,27,28-テトラキス（エトキシカルボニル）-メトキシ-カリックス[4]アレン18.4mg（18.4 μ mol）、色素として2,6-ジプロモ-3'-メトキシインドフェノール7.1mg（18.4 μ mol）を溶解し、これを透明ポリプロピレンシート上にドクターブレードにて600 μ m厚に塗布した。室温中で放置し乾燥させると、徐々に濁度が上がり、最終的に茶白色の膜状物を得た、この膜状物を透明ポリプロピレンシートと共に1cm角に切断し、茶白色の膜状物が透明ポリプロピレンシートに固着したイオン測定具を得た。

【0065】実施例6

実施例5にて作製したイオン測定具を第2図に示す反射光量測定装置に装着し、各種塩化ナトリウム濃度の2-モルホリノエタンスルホン酸10mM緩衝液（pH5.5に水酸化カリウムにて調製）25 μ lを滴下した。波長670nmでの反射光量を測定し、塩化ナトリウム濃度0mMの時の反射光量を基準として吸光度を計算した。結果を第5図に示した。図5から、ナトリウムイオンを10 \sim 0.1mMの間で測定出来ることがわかる。また一定の反射光量になるまでに要した時間をも測定した。要した時間はいずれも15 \sim 20秒の範囲内であった。

【0066】比較例4

テトラヒドロフラン10mlに、非水溶性高分子としてポリ塩化ビニル（平均重合度1000）300mg、有機溶剤としてo-ニトロフェニルオクチルエーテル600mg、ナトリウムイオノフォアとして5,11,17,23-テトラ- ϵ -ブチル-25,26,27,28-テトラキス（エトキシカルボニル）-メトキシ-カリックス[4]アレン18.4mg（18.4 μ mol）、色素として2,6-ジプロモ-3'-メトキシインドフェノール7.1mg（18.4 μ mol）を溶解し、これを透明ポリプロピレンシート上にドクターブレードにて600 μ m厚に塗布した。室温中で放置し乾燥させ、測定具を均一な透明褐色の膜状物として得た。

【0067】このようにして製造した測定具を第2図に示す反射光量測定装置に装着し、各種塩化ナトリウム濃度の2-モルホリノエタンスルホン酸10mM緩衝液（pH5.5に水酸化カリウムにて調製）25 μ lを滴下した。波長670nmでの反射光量を測定したが、3分経過しても一定の反射光量に到達しなかった。応答速度の遅く実施例6と同じ条件では測定を行うことはできなかった。

【0068】実施例6および比較例5から本発明のイオン測定具を用いて迅速に測定を行うことができることが示された。

【0069】比較例5 \sim 10、実施例7 \sim 227

表2に示す非水溶性高分子、有機溶剤、水溶性高分子、実施例5と同一のナトリウムイオノフォア、色素を用い、実施例5と同様の操作にて、ナトリウムイオン用のイオン測定具を作製した。作製した測定具を第2図に示す反射光量測定装置に装着し、塩化ナトリウム濃度10mMの2-メルホリノエタンスルホン酸10mM緩衝液(pH5.5に水酸化カリウムにて調製)25 μ lを滴下した。波長670nmでの反射光量を測定し、そのときの反射光量が一定にな*

*るまでの時間を測定した。結果を表2に示した。実施例5~10よりわかるように、非水溶性高分子、有機溶剤、水溶性高分子のいずれかか欠けても迅速な応答は得られなかった。また、実施例7~22より各種非水溶性高分子、有機溶剤、水溶性高分子の組み合わせにおいて迅速な応答が得られることが示された。

【0070】

【表2】

表 2

	非水溶性高分子	有機溶剤	水溶性高分子	応答時間(秒)
比較例5	なし	セバシン酸ジオクチル	ポリエチレングリコール	180秒以上
比較例8	"	フタル酸ジオクチル	ポリエチレングリコール	180秒以上
比較例7	"	"	ポリビニルピロリドン	180秒以上
比較例8	"	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	180秒以上
比較例8	ポリ塩化ビニル	なし	ポリエチレングリコール	反射量変化なし
比較例10	"	セバシン酸ジオクチル	なし	180秒以上
実施例7	"	"	ポリビニルピロリドン	17
実施例8	"	"	ポリエチレングリコール	18
実施例8	"	フタル酸ジオクチル	ポリエチレングリコール	20
実施例10	"	"	ポリビニルピロリドン	20
実施例11	"	o-ニトロフェニルオクチルエーテル	ポリエチレングリコール	16
実施例12	"	"	ポリビニルピロリドン	18
実施例13	ポリメタクリル酸メチル	セバシン酸ジオクチル	ポリエチレングリコール	18
実施例14	"	"	ポリビニルピロリドン	18

【0071】

【表3】

*【図面の簡単な説明】

【図1】 本図は実施例1にて作製された物質測定具をイオン交換水で洗浄した後の有機相の構造を電子顕微鏡で写真撮影した写真である。

【図2】 実施例、比較例にて用いた反射光量測定装置の構成を示す図である。

【図3】 実施例4において測定した亜鉛イオン濃度と測定された吸光度の関係を示す図である。

【図4】 比較例3において測定した亜鉛イオン濃度と測定された吸光度の関係を示す図である。

【図5】 実施例6において測定したナトリウムイオン濃度と測定された吸光度の関係を示す図である。

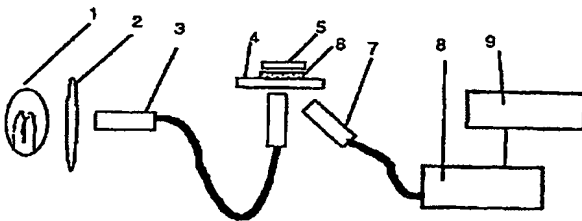
【符号の説明】

- 1 キセノンランプ
- 2 集光レンズ
- 3 投光用光ファイバー
- 4 台（石英板）
- 5 白色反射板
- 6 本発明の物質測定具
- 7 受光用光ファイバー
- 8 瞬間マルチ測光装置（大塚電子株式会社製 MCPD-10 00型）
- 9 データ処理装置（大塚電子株式会社製）

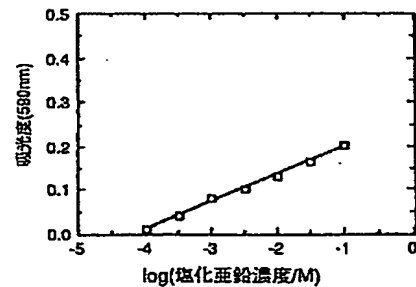
表 2 (続き)

	非水溶性高分子		水溶性高分子	応答時間 (秒)
実施例 15	ポリメタクリル酸メチル	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 16	〃	〃	ポリビニルピロリドン	20
実施例 17	〃	6-ニトロフェニルオクチルエーテル	ポリエチレングリコール	17
実施例 18	ポリスチレン	セバシン酸ジオクチル	ポリエチレングリコール	18
実施例 19	〃	〃	ポリビニルピロリドン	19
実施例 20	〃	フタル酸ジオクチル	ポリエチレングリコール	20
実施例 21	〃	〃	ポリビニルピロリドン	18
実施例 22	〃	6-ニトロフェニルオクチルエーテル	ポリエチレングリコール	18

【図2】

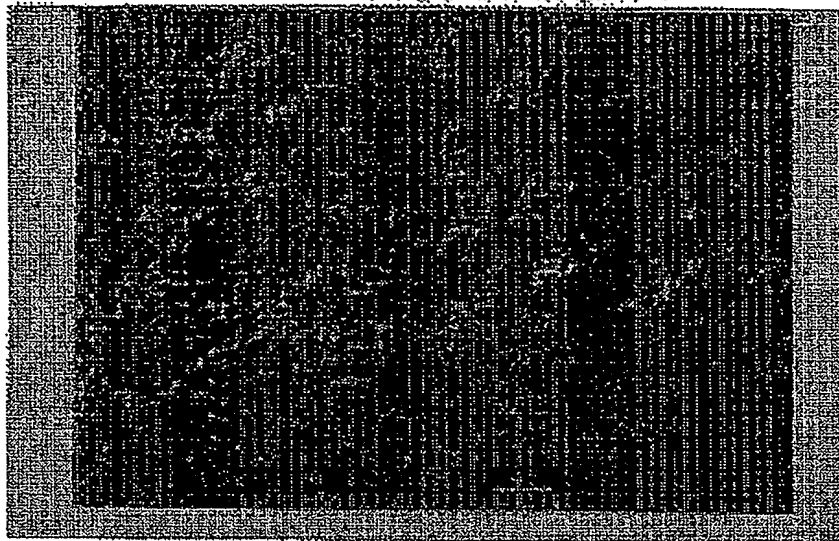


【図3】



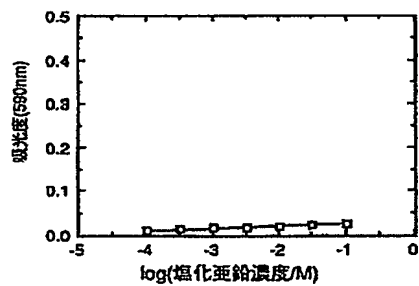
【図1】

図面代用写真



写真

【図4】



【図5】

